Ultrafast breaking of spatial/time reversal symmetry by a single-cycle light-field in strongly correlated chargers/spins

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Recent advances of ultrafast laser technologies and quantum materials open non-perturbative manipulations of correlated electron systems [1-3]. Thus, a strong light-field enables us to expect realization of a new class of photoinduced symmetry breaking which is different from the conventional regime, i.e., breaking of space inversion symmetry in ferroelectric compounds and that of time reversal symmetry in antiferromagnets. Here, we would like to discuss i) second harmonic generation (SHG) induced by non-dissipative light-currents in a centrosymmetric superconductor and ii) ultrafast helicity-dependent polarization rotation in a Kitaev-type spin liquid candidate α-$RuCl_3$ as new examples of photoinduced symmetry breaking.

i) An electromagnetic oscillation of light cannot directly access to space inversion symmetry breaking because of its symmetric nature on the time axis (i. e., the time average of the oscillation is zero). However, recent developments of ultrashort laser technologies enable us to control the direction of charge motion by carrier-envelope phase (CEP) control of a strong light field. Considering non-perturbative and non-dissipative light-matter interactions during an ultrashort pulse, we can expect petahertz control of the space inversion symmetry in solids. Here, in a layered organic superconductor, SHG is observed by using a single-cycle 6 femtosecond near infrared pulse, which is in contrast to the perturbation theory where even harmonics are forbidden in centrosymmetric systems. The SHG shows a CEP sensitive nature and an enhancement near the superconducting temperature. The result and its quantum many-body analysis indicate that a polarized current is induced by non-dissipative acceleration of charge, which is amplified by superconducting fluctuations.

ii) A zigzag type AF order ($T<T_N$=7 K) in a layered honeycomb lattice Mott insulator α-$RuCl_3$ with a large spin-orbit coupling attracts much attention. In this compound, a Kitaev-type spin-liquid state is expected to be realized because of its characteristic electronic and structural properties, i. e., bond-dependent anisotropic exchange interactions between J_{eff}=1/2 pseudospins on the honeycomb lattice precludes a long-range order for $T> T_N$. Moreover, the Majorana fermion nature and its possible application to a topological quantum computer are of much interest. Here, we report that an ultrafast helicity-dependent polarization rotation is induced by a circular polarized pump light. Immediate (< 0.1 ps) responses of rise and decay within time resolution are followed by a 400 fs decay at the spin-liquid ($T>T_N$) phase. In addition, a helicity-independent 10 ps growing-up component is also observed only at the AFM phase. The ultrafast (<0.1 ps, 400 fs) and slower (10 ps) components respectively reflect the spin dynamics in the spin liquid and that in the long range order.

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