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Normal mode selectivity in ultrafast Raman excitations in C_{60}[1]

GUOPING ZHANG, Indiana State University, THOMAS F. GEORGE, University of Missouri-St. Louis, ZHANG-GEORGE TEAM — Ultrafast Raman spectra are a powerful tool to probe vibrational excitations, but inherently they are not normal-mode specific. For a system as complicated as C_{60}, there is no general rule to target a specific mode. A detailed study presented here aims to investigate normal mode selectivity in C_{60} by an ultrafast laser. To accurately measure mode excitation, we formally introduce the kinetic energy-based normal mode analysis which overcomes the difficulty with the strong lattice anharmonicity and relaxation. We first investigate the resonant excitation and find that mode selectivity is normally difficult to achieve. However, for off-resonant excitations, it is possible to selectively excite a few modes in C_{60} by properly choosing an optimal laser pulse duration, which agrees with previous experimental and theoretical findings. Going beyond the phenomenological explanation, our study shines new light on the origin of the optimal duration: The phase matching between laser field and mode vibration determines which mode is strongly excited or suppressed. This finding is very robust and may be a useful guide for future experimental and theoretical studies in more complicated systems.

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