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Title

Nonlinear phononics as an ultrafast route to lattice control

Source

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Abstract

Two types of coupling between electromagnetic radiation and a crystal lattice have so far been identified experimentally. The first is the direct coupling of light to infrared-active vibrations carrying an electric dipole. The second is indirect, involving electron-phonon coupling and occurring through excitation of the electronic system; stimulated Raman scattering(1-3) is one example. A third path, ionic Raman scattering (IRS; refs 4,5), was proposed 40 years ago. It was posited that excitation of an infrared-active phonon could serve as the intermediate state for Raman scattering, a process that relies on lattice anharmonicities rather than electron-phonon interactions(6). Here, we report an experimental demonstration of IRS using femtosecond excitation and coherent detection of the lattice response. We show how this mechanism is relevant to ultrafast optical control in solids: a rectified phonon field can exert a directional force onto the crystal, inducing an abrupt displacement of the atoms from their equilibrium positions. IRS opens up a new direction for the optical control of solids in their electronic ground state(7-9), different fromcarrier excitation(10-14).