

Good vibrations feel the Force

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Strong-field mid-infrared excitation allows to drive lattice vibrations of a crystal into the highly anharmonic regime. Here, the atoms oscillate not only at their fundamental frequency but also at overtones, so called higher harmonics. The measurement of this atomic motion far away from equilibrium allows to reconstruct the interatomic potential. Credit: Max Planck Institute for the Structure and Dynamics of Matter

A group of researchers led by Andrea Cavalleri at the Max Planck Institute for Structure and Dynamics of Matter (MPSD) in Hamburg has demonstrated a new method enabling precise measurements of the interatomic forces that hold crystalline solids together. The paper Probing the Interatomic Potential of Solids by Strong-Field Nonlinear Phononics, published online in *Nature*, explains how a terahertz-frequency laser pulse can drive very large deformations of the crystal. By measuring the highly unusual atomic trajectories under extreme electromagnetic transients, the MPSD group could reconstruct how rigid the atomic bonds are at large distances from the equilibrium arrangements. This promises new insights into the mechanical properties of matter and their instability near phase changes.

Crystals are held together by extremely strong forces, which determine all their thermal and [mechanical properties](#). The temperature at which a specific material melts or changes shape and the material's resistance to pressure and shear distortions are all determined by this 'force field'. It is the basis of any textbook description of a material and is routinely calculated by sophisticated theoretical methods. Still, until now no experiment could quantitatively validate these calculations or at least measure the [force](#) field.

In a recent study by the MPSD group led by Andrea Cavalleri, ultrashort laser flashes at mid-infrared frequencies were used to move atoms far away from their equilibrium arrangement. By measuring how the same atoms were made to ring after the impulse had been turned off, the MPSD research group could reconstruct the nature of the forces that hold the crystal together.

"We use strong laser fields to drive the atoms to displacements where their dynamics can no longer be described within the harmonic approximation," explains Alexander von Hoegen, doctorate at the MPSD and first author of this paper. "In this situation, the restoring forces

acting on the atoms are no longer linear proportional to the displacements from the equilibrium positions, as they would be in the case of small oscillations in a pendulum."

Such nonlinear phononics is for example manifested by the fact that the atoms not only oscillate at their natural frequency, but also at multiple overtones, the so-called higher harmonics observed in this study.

The corresponding atomic displacements, enormous on the scale of the interatomic distances, are nevertheless only of the order of a few picometers, that is a millionth of a billionth of a meter. The vibrations were traced with a second, even shorter [laser](#) pulse. Although the atoms were found to oscillate with speeds beyond 1000 m/s, their motion could be traced in ultra-slow motion. This time-resolved measurement was key to reconstructing the forces acting on the [atoms](#).

This work by the MPSD establishes a new type of nonlinear spectroscopy that captures one of the most fundamental microscopic properties of materials, underscoring the power of new advanced optical sources and paving the way to a future, even more insightful class of experiments at the Hamburg X-ray Free Electron Laser.

More information: Probing the interatomic potential of solids with strong-field nonlinear phononics, A. von Hoegen, R. Mankowsky, M. Fechner, M. Först & A. Cavalleri, *Nature*, online, 21. Feb. 2018, [DOI: 10.1038/nature25484](https://doi.org/10.1038/nature25484)

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