Supplementary Information for

Giant Lattice Expansion by Quantum Stress and Universal Atomic Forces in Semiconductors under Instant Ultrafast Laser Excitation

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1. Excitation Deformation Potential and Bonding Electronic Density

As discussion in the main text, the excitation stress can have different magnitude in various materials. The ability to produce the stress (in the same intensity for various semiconductors) is related to its excitation deformation potentials ($dE_g^{'} / d\varepsilon$). Here, $E_g^{'}$ is different from the normal band gap of E_g because the quasi Fermi levels E_{Fe}, E_{Fh} can be shifted into the conduction band and the valence band under a fs excitation with a typical intensity, such as 10%. As such, $dE_g^{'} / d\varepsilon \neq dE_g / d\varepsilon$ under a large excitation. However, if the excitation intensity is small, the case will be close to that in ground state. For example, the ground-state deformation potential of GST has been reported to be abnormal (i.e. $dE_g / d\varepsilon > 0$) [Ref. S1]. Therefore, the stress under a relatively low excitation (such as at 3%) is positive [Figure 2 (a) in the main text]. However, the stresses under the large excitation (> 6%) change to the normal negative one and lead to the lattice expansion.

On the other hand, it is straightforward that the excitation deformation potential is related to the bonding strength. Thus, we define a bonding electronic density ($\rho_{eb}=\frac{N_{eb}}{CN \cdot BL}$) to reflect the strength. Here, N_{eb} is the average number of electrons per atom for bonding, CN is the average coordination number, and BL is the average bond length. Figure 1(c) displays there is a linear dependence of the excitation deformation potential on the bonding electronic density for different materials. Thus, the deformation potential is material specific and we can estimate an excitation stress in various materials by a simple analysis on their bonding strength.

2. Evaluation of the volume variation responding to excitation induced stress.

The change of volume responding to excitation is calculated by changing the volumes to release the stresses (Figure S1 is an example for diamond with a 6% excitation). Here, the volumes with zero stresses is the final relaxed volume under excitation.

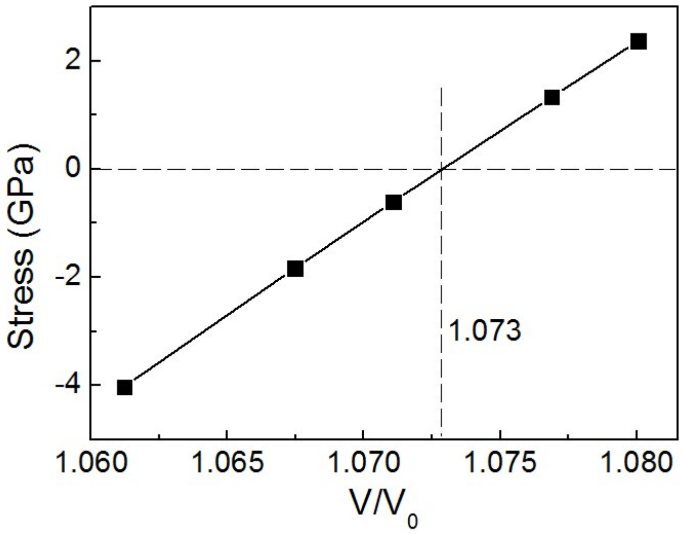

Figure S1. The relationship between stress and volume under 6% excitation. When the V/V_0 is 1.073, the stress is zero. As such, the volume will expand by 7.3% to response to 6% excitation.

References

[S1] B. Xu, Y. Su, Z. G. Liu, C. H. Zhang, Y. D. Xia, J. Yin, Z. Xu, W. C. Ren, and Y. H. Xiang, Appl. Phys. Lett. **98,** 142112 (2011).