Molecular Dynamics Simulations Demonstrate Laser Healing of CNT Defects

The electronic properties of single-wall carbon nanotubes (CNTs) can be severely impaired by common structural imperfections, such as the pentagon–heptagon pair defect caused by a 90° rotation of a carbon dimer (a Stone–Wales-type transformation). Methods developed for the repair of such defects are therefore important for the pragmatic fabrication of CNTs in electronic devices. To this end, A.H. Romero of CINVESTAV, Queretaro, Mexico; M.E. Garcia of Universität Kassel, Germany, CINSaT; F. Valencia and colleagues at IPICYT, San Luis Potosí, Mexico; and H.O. Jeschke of Johann Wolfgang Goethe-Universität, Frankfurt/Main, Germany, have demonstrated with computer simulations that femtosecond laser pulses can repair atomic-scale defects in nanostructures.

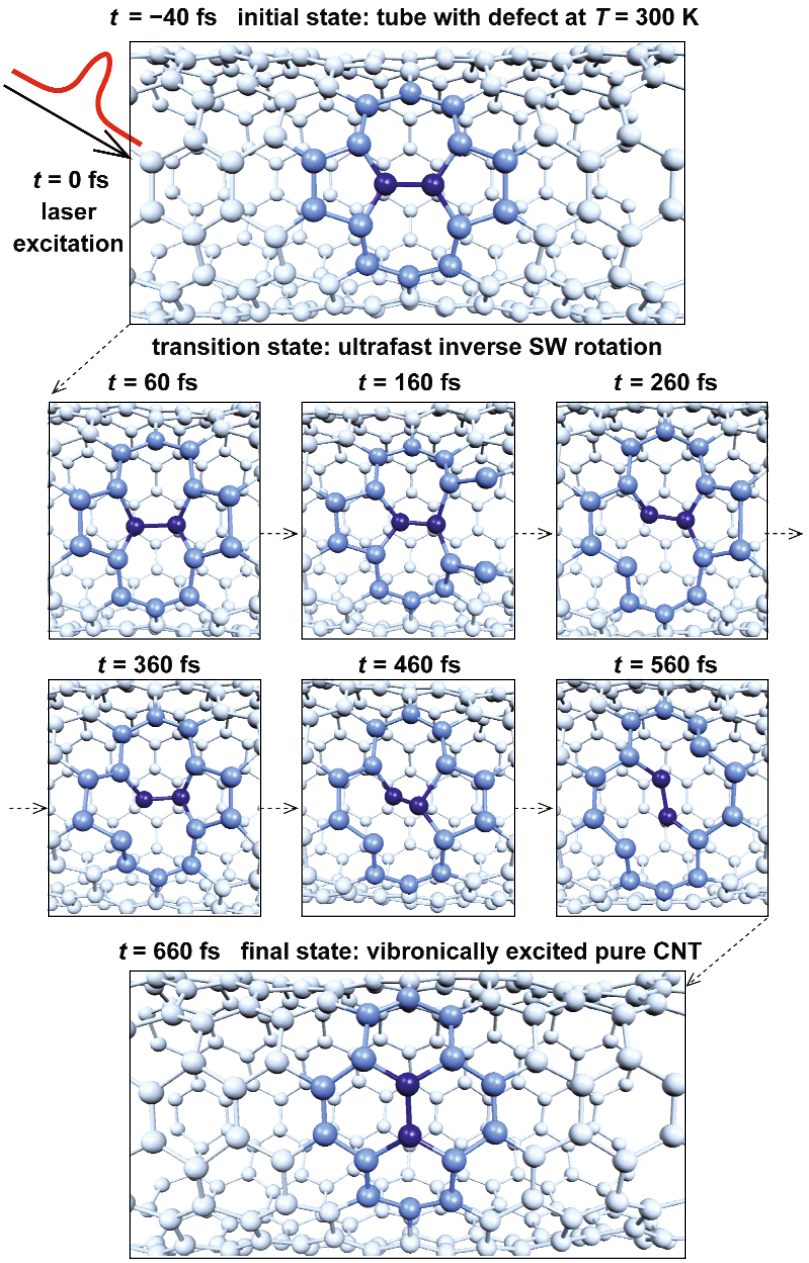

Figure 1. Simulated snapshots of the elimination of a pentagon–heptagon defect in a (7,7) carbon nanotube due to illumination with an ultrashort laser pulse. The absorbed energy induces bond-breaking processes primarily in the defect region (highlighted with darker color), making the rotation of a carbon dimer possible. Reprinted with permission from NanoLetters **5** *(2005), p. 1361; DOI: 10.1021/nl050626t. © 2005 American Chemical Society.*

As reported in the July 13 issue of *NanoLetters* (p. 1361; DOI: 10.1021/nl050626t), the researchers performed non-adiabatic tight-binding molecular dynamics simulations on (7,7) zigzag and (12,0) armchair CNTs. Using previously published parameters for this method, the only variables were the energy absorbed per atom and the type of CNT. Periodic boundary conditions were employed to simulate infinitely long CNTs with a defect density of 3%. Laser-induced structural changes were determined from simulating the dynamics of the atoms on the potential energy surface (PES), defined in terms of the electronic free energy, entropy, and temperature. Shown in Figure 1 is the simulated inversion of the generalized Stone–Wales transformation. After equilibrating the CNTs at 300 K, an ultrashort laser pulse, with a 50 fs duration and a central quantum energy of 1.96 eV, excited the system—that is, it changed the occupations of the electrons—resulting in the generation of a coherent phonon corresponding to a large-amplitude radial breathing mode during the first 400 fs. Afterwards, the PES changed by the laser pulse caused the defect's two central atoms to break their bonds with adjacent atoms. The dimer then underwent a complex motion—one component perpendicular to the CNT surface and another in the pseudo-plane of the CNT—that healed the defect.

The researchers demonstrated the universality of this mechanism by simulating the defect healing with different laser-pulse parameters and by inducing an inverse Stone–Wales transformation in a graphene sheet. The researchers said, however, that this type of dimer rotation cannot occur in C_{60}.

The researchers determined that in all their simulations, the excitation threshold for defect repair corresponded to an excitation of 7% of the valence electrons. They said that the laser pulses used to heal defects should be chosen below the laser damage threshold, which should be determined experimentally because it varies considerably with tube diameter and chirality.

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