Four-Phonon Scattering Reduces Intrinsic Thermal Conductivity in Graphene and the Contributions from Flexural Phonons

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We have developed a formalism of the exact solution to the linearized phonon Boltzmann transport equation (BTE) for thermal conductivity calculations including three- and four-phonon scattering. We find strikingly high four-phonon scattering rates in single-layer graphene (SLG) based on the optimized Tersoff potential. The reflection symmetry in graphene, which forbids the three-ZA (out-of-plane acoustic) scattering, allows the four-ZA processes. As a result, the large phonon population of the low-energy ZA branch originated from the quadratic phonon dispersion leads to very high four-phonon scattering rates, even much higher than the three-phonon scattering rates at room temperature. These four-phonon processes are dominated by the normal processes, which lead to a failure of the single mode relaxation time approximation. Therefore, we have solved the exact phonon BTE using an iterative scheme and then calculated the length- and temperature-dependent thermal conductivity. We find that the predicted thermal conductivity of SLG is significantly lower than the previously predicted value from three-phonon scattering only. The relative contribution of the ZA branch is reduced from 70 % to 30 % when four-phonon scattering is included, consistent with molecular dynamics simulations. Furthermore, we have demonstrated that four-phonon scattering in multi-layer graphene and graphite is not strong due to the ZA splitting by the interlayer van der Waals interaction. We also demonstrate that the five-phonon process in SLG is not strong due to the reflection symmetry.