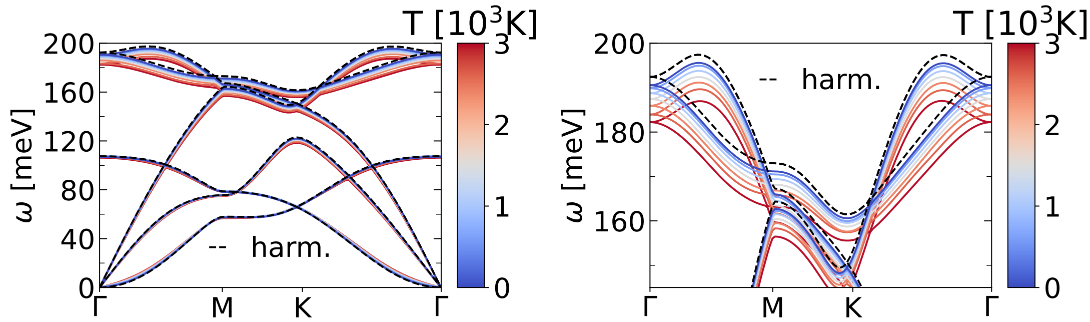
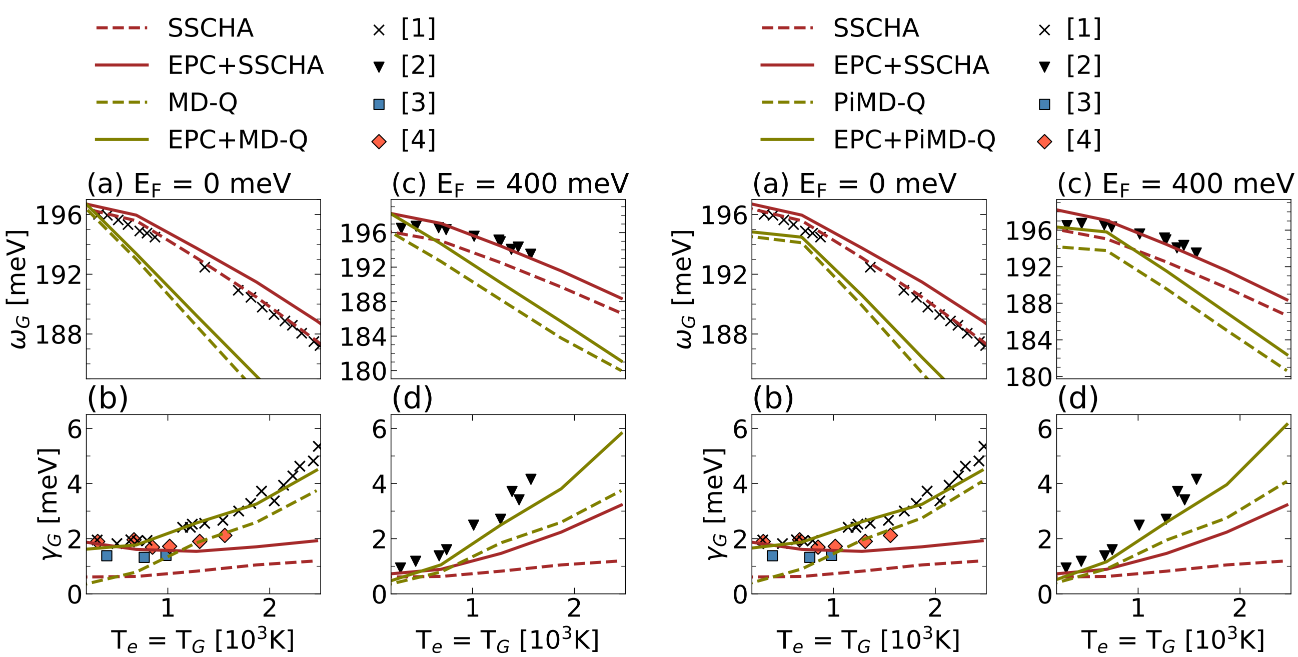
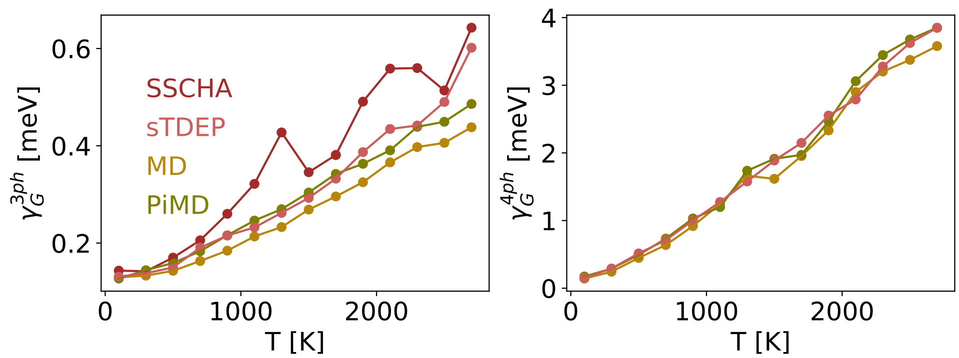


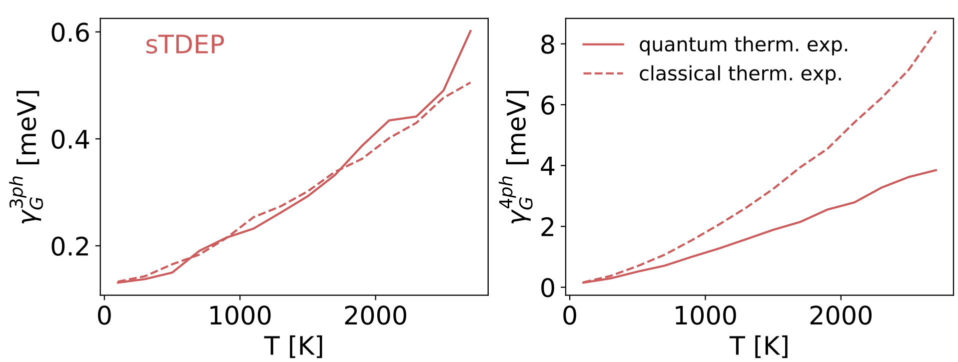
Supplementary Figure 1. Graphene phonon dispersion for various temperatures as obtained from SSCHA. Right panel is a close-up of the left panel, highlighting the modifications of the relevant optical phonons.



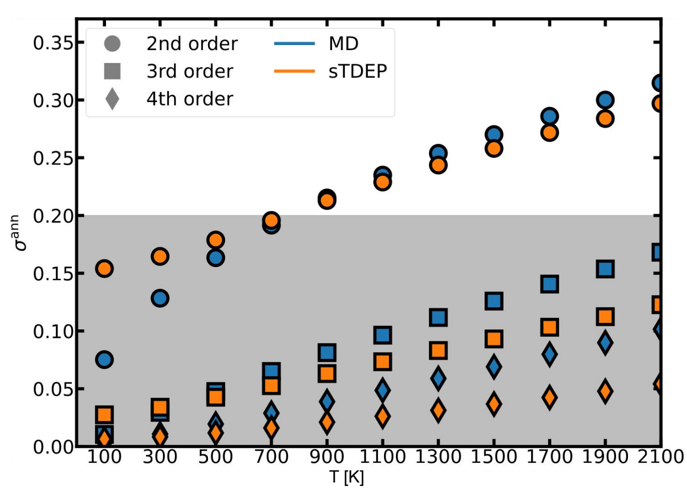
Supplementary Figure 2. For the two different doping regimes, we show the total EPC and anharmonicity induced G mode frequency renormalization (a,c) and linewidth (b,d) on the smaller temperature range. We compare anharmonic effects obtained with TDEP with MD (left panels) and PiMD (right panels) and with SSCHA. (a,c) Anharmonic effects have a prevailing temperature dependence at smaller temperatures shown here and determine the G mode frequency decrease with temperature observed in experiments [1, 2]. (b,d) Linewidth increase with temperature observed in Ref. [1–4].



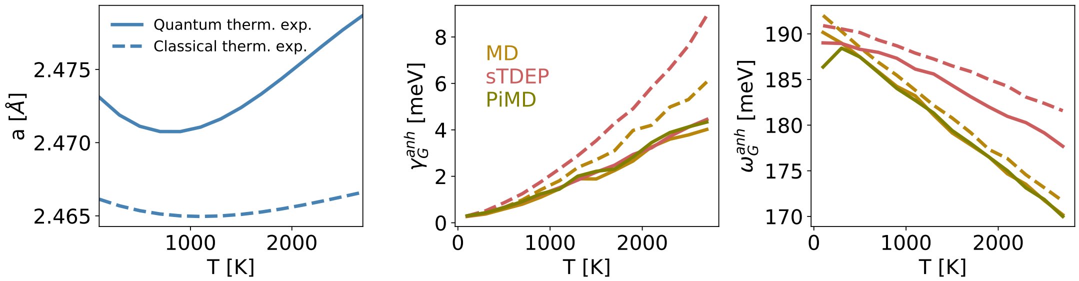
Supplementary Figure 3. Comparison between three- and four- phonon linewidths. In the first panel, we compare a three-phonon anharmonic contribution to the linewidth as obtained from SSCHA, and from TDEP using MD and canonical sampling. In the second panel, we show a much larger four-phonon contribution obtained from TDEP.



Supplementary Figure 4. Comparison between a three- and four-phonon contribution to the linewidth with classical and quantum thermal expansion as obtained from sTDEP. The difference between the quantum and classical thermal expansion effects are only noticeable in the four phonon contribution to the linewidth.



Supplementary Figure 5. Evolution of the anharmonicity measure [5] at the second, third and fourth order computed with MD and sTDEP. The low values, below the 0.2 threshold suggested in [5], indicate an accurate reproduction of the forces.



Supplementary Figure 6. In the first panel, we show the thermal expansion of the lattice constant, calculated using a quantum and classical approach in the free energy calculation. The second and third panels show a comparison between TDEP results obtained with MD, PiMD and SCHA for the G mode linewidth and frequency, respectively. The lattice expansion is shown in full lines for quantum and dashed lines for classical dynamics. Using the lattice constant obtained in a classical way, the linewidth is overestimated, while the frequency temperature dependence only undergoes a rigid shift. The overlap between PiMD and MD results indicates that quantum fluctuations are only important at very low temperatures. The largest quantum effect is the lattice expansion.

Supplementary References

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