Supporting Information for:

In-Plane Breathing and Shear Modes in Low-Dimensional Nanostructures

Dan Liu,[1] Colin Daniels,[2] Vincent Meunier,[2] Arthur G. Every,[3] and David Tománek[1,*]

[1] *Physics and Astronomy Department, Michigan State University, East Lansing, Michigan 48824, USA*
[2] *Department of Physics, Applied Physics, and Astronomy, Rensselaer Polytechnic Institute, Troy, NY 12180, USA*
[3] *School of Physics, University of the Witwatersrand, Private Bag 3, 2050 Johannesburg, South Africa*

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I. COMPARISON BETWEEN SCALING LAWS FOR THE RADIAL-LIKE BREATHING MODE IN GRAPHENE NANORIBBONS

In the main manuscript, we have derived a scaling law for ω_{RBLM} as a function of nanoribbon width W based on continuum elasticity theory for 2D systems

$$\omega_{RBLM} = \frac{c_{RBLM}}{W + \delta W} \tag{S1}$$

$$\text{with} \quad c_{RBLM} = \pi \sqrt{\frac{c_{11}}{\rho_{2D}}} \,.$$

For graphene nanoribbons (GNRs), we use [1] $c_{11} = 352.6$ N/m, and $\rho_{2D} = 0.743 \times 10^{-6}$ kg/m^2. The effect of edge termination, which may vary from sample to sample, is represented by the parameter δW. We found that $\delta W = 1.8$ Å describes H-termination properly.

We note that the functional dependence of ω_{RBLM} on the nanoribbon width W in Eq. (S1) differs significantly from previously used scaling laws covering the entire range from narrow to wide GNRs. Several authors have used the expression [2–4]

$$\omega_{RBLM} = \frac{a}{\sqrt{W}} + b \,. \tag{S2}$$

The values of the parameters a and b for GNRs are very similar in References 2–4. Parameters obtained from fits to density functional theory (DFT) calculations of Ref. 3 are $a = 1667.9$ cm^{-1}Å$^{1/2}$ and $b = -210.2$ cm^{-1}. With these parameters, ω_{RBLM} becomes negative and unphysical in wide GRNs.

An alternative expression [5]

$$\omega_{RBLM} = \frac{c}{W} + \frac{d}{\sqrt{W}} + e \tag{S3}$$

has been proposed to alleviate the incorrect behavior of ω_{RBLM} in Eq. (S2) for wide GNRs. The parameters used in Ref. 5 for GNRs are $c = 1584.24$ cm^{-1}Å, $d = 351.98$ cm^{-1}Å$^{1/2}$ and $e = -10.00$ cm^{-1}. Yet also with this scaling law, ω_{RBLM} eventually becomes negative in ultra-wide GNRs.

The dependence of ω_{RBLM} in hydrogen-terminated GNRs of width W, described by Eqs. (S1), (S2) and (S3), is compared for different width ranges in Fig. S1 and experimental data.

As seen in Fig. S1(a), all expressions correctly predict a monotonic decrease of ω_{RBLM} with increasing GNR width W in narrow GNRs. There is, however, a quantitative difference of up to $\lesssim$20% between the results for

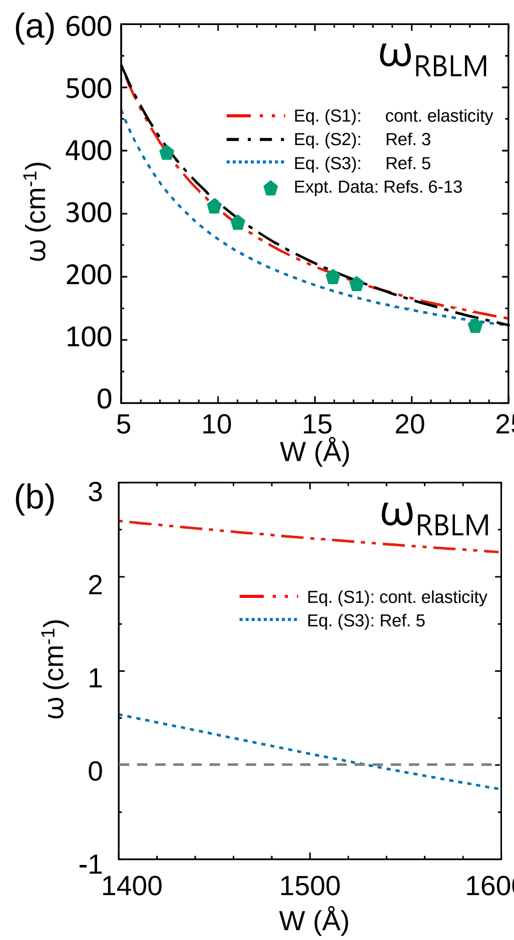

FIG. S1. Dependence of ω_{RBLM} on the GNR width W according to expressions in Eqs. (S1), (S2) and (S3). Numerical results are presented for W in the range of (a) $5-25$ Å and (b) 1400 Å$-$1600 Å.

* E-mail: tomanek@pa.msu.edu

narrow GNRs with $5 \lesssim W \lesssim 25$ Å. Experimental data [6–13] agree best with the scaling laws in Eqs. (S1) and (S2).

Qualitative differences between the different expressions become more obvious in wide GNRs with 1400 Å$\lesssim W \lesssim$1600 Å according to Fig. S1(b). Whereas ω_{RBLM} remains positive and correctly approaches zero for $W \to \infty$ according to Eq. (S1), the frequency becomes negative for $W > 63$ Å according to Eq. (S2) and for $W > 1530$ Å according to Eq. (S3). Asymptotically, for $W \to \infty$, $\omega_{RBLM} = -210.2$ cm^{-1} according to Eq. (S2) and $\omega_{RBLM} = -10$ cm^{-1} according to Eq. (S3).

A $1/W$ functional dependence of ω_{RBLM} with the correct asymptotic behavior $W \to \infty$ has been discussed previously [5, 14–16] with the conclusion that it only may reproduce observed data in "not too narrow nanoribbons" [16]. We also note that the zone-folding approach used in Refs. 14–16 can not easily accommodate edge effects in narrow GNRs. Among the proposed functional dependencies of ω_{RBLM} on W, only Eq. (S1) covers the entire range from narrow to wide nanoribbons.

II. ATOMIC DISPLACEMENTS IN RADIAL BREATHING MODES OF CIRCULAR GRAPHENE DISCS

The structure of different hydrogen-terminated graphene discs is represented by ball-and-stick models in Fig. S2. The disc radius R is defined as the distance from the center to the outermost carbon atom. We consider two discs, identical to those described in Fig. 2(d) of the main manuscript, and display a snapshot of the atomic motion during the radial breathing mode. We found no notable difference in the character of the breathing modes between results obtained by DFT and the REBOII force fields used in the main manuscript.

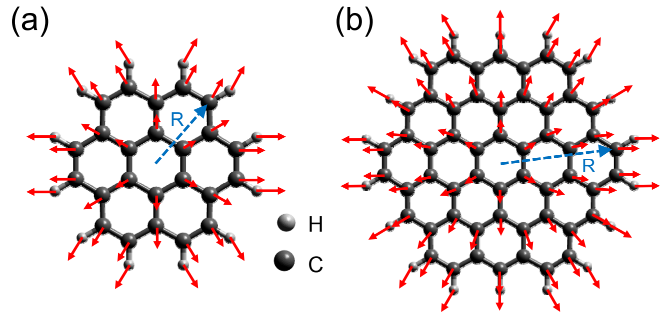

FIG. S2. Atomic structure of hydrogen-terminated circular graphene discs with the radii (a) $R = 3.75$ Å, (b) $R = 6.20$ Å. Atomic motion during the radial breathing motion, based on DFT calculations, is depicted by displacement vectors that are shown by the red arrows.

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