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Lepetit and Jackson Reply

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Lepetit and Jackson Reply: We agree with Clougherty's Comment [1] about the existence of a singularity at zero phonon frequency which appears in the coupling term used in Ref. [2] to model the interaction between an incoming hydrogen atom and a suspended graphene layer. This singularity exists only in the continuum case (a suspended graphene sheet of infinite size). In our model, we use a finite sampling of the first Brillouin zone (1BZ), reducing the graphene sheet to finite size and regularizing this singularity with a low frequency cutoff. Results shown in Ref. [2] were obtained with a 191 GHz (0.79 meV) cutoff, corresponding to a sampling of the $\Gamma - M - K$ triangle of the 1 BZ with 2000 phonon modes.

The question then turns into one about the influence of this cutoff on the sticking probability. In our study, we consider thermal energy collisions, where sticking is a fast process occurring typically at a time scale of the order of a picosecond or smaller (i.e., with frequencies higher than a terahertz). We can anticipate that changes in the low frequency part of the phonon spectrum below this threshold should have little impact on the physical process. This was checked by comparing the sticking probability of Ref. [2] (Fig. 3, suspended case) with a more accurate one involving a 114 GHz low frequency cutoff and a denser sampling of the $\Gamma - M - K$ triangle involving 5000 phonon modes. Results differ by less than 1% for energies below than 20 meV, except in two energy bands, approximately 2 meV wide around 7 and 15 meV, where differences can reach 10%. For most collision energies, sticking is essentially a fast direct process, involving phonon excitations with energies $E - E_i$ resonant with the transitions from the initial collisional continuum state with positive energy E (in the range 1–20 meV) down to stuck bound states with negative energy E_i (5 states with energies from –25 meV for the ground stuck state to –0.5 meV for the highest energy one considered; see Table 1 in Ref. [3]). These phonon energies are well above the ones of the cutoff, and sticking is unaffected by changes in the low frequency part of the phonon spectrum. However, for energies near 7 and 15 meV, the trapping mechanism described in [2–4] takes place. The incoming hydrogen is trapped close to the surface in a diffraction-mediated selective adsorption resonance that has more time to exchange low frequency phonons with the surface before being stuck. In this case, a change in the cutoff frequency has some visible effect on the sticking probability. However, as this indirect mechanism contributes less than the direct one to sticking, the effect of the cutoff change on global sticking remains limited.

Further lowering the cutoff frequency in our calculation to describe larger size membranes would lead to an extremely dense first Brillouin zone sampling not accessible with our current computing facilities, and would, in

any case, require a higher-order treatment of the coupling as suggested by Clougherty. Within the frame of a variational mean-field theory [1,5,6], he finds a critical energy below which the sticking of incident hydrogen atoms vanishes. The numerical value for this critical energy is derived for a model system which differs significantly from ours. His model Hamiltonian describes a single continuum state coupled to a single bound one (energy –25 meV) on an uncorrugated membrane. In our model, the full continuum is coupled to the 5 bound states already mentioned, and the critical energy is dependent on these bound state energies. The corrugation included in our model was shown in our Letter to significantly impact the sticking. His formalism gives a sticking probability of zero in the continuum limit of an infinitely sized membrane with a temperature less than 1 K [1], whereas in our calculation, our finite size graphene sample was at 10 K. It would be interesting to know how these differences impact the critical energy evaluation, and, eventually, in the ultralow energy range where reflection occurs on suspended graphene, how differently supported graphene, which has a low frequency cutoff in the meV energy range due to the relative graphene-substrate motion, would behave.

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- [1] D. P. Clougherty, preceding Comment, *Phys. Rev. Lett.* **113**, 069601 (2014).
- [2] B. Lepetit, and B. Jackson, *Phys. Rev. Lett.* **107**, 236102 (2011).
- [3] B. Lepetit, D. Lemoine, Z. Medina, and B. Jackson, *J. Chem. Phys.* **134**, 114705 (2011).
- [4] Z. Medina and B. Jackson, *J. Chem. Phys.* **128**, 114704 (2008).
- [5] Y. Zhang and D. P. Clougherty, *Phys. Rev. Lett.* **108**, 173202 (2012).
- [6] D. P. Clougherty and Y. Zhang, *Phys. Rev. Lett.* **109**, 120401 (2012).