

## Strong slip-induced anomalous enhancement and redshifts in wide-range optical absorption of graphite under uniaxial pressure

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Ji-Chang Ren,<sup>abc</sup> Rui-Qin Zhang,<sup>\*a</sup> Zejun Ding<sup>\*b</sup> and Michel A. Van Hove<sup>d</sup>

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### Methods discussions

The approach we used is the density-functional perturbation theory based on GGA. It is reasonable to apply this method to our system, because of the following reasons. As the interlayer distance decreases, the two dimensional character of the graphene layer tends to disappear. Thus, the long range correlation induced by long range dispersion of electron density between layers can be ignored or can be cancelled by the corresponding exchange interaction. In the process of compression, since the interlayer bond has not been formed, the in-plane  $\pi$  orbitals retain and thus the electron density is not so much localized to cause noticeable self-interaction error and correlation interactions. Thus, although the approach adopted in our work is at the GGA level, it provides an acceptable accuracy to demonstrate the redshifts of the peak C. In order to confirm our results, we have redone the calculations with the inclusion of a local field effect (LFE) from PBE-GGA functionals and random phase approximation (RPA). The RPA is a suitable method to describe the long range correlation effect. The LFE is the correction for the system with inhomogeneous density, superior in describing the electron density dispersion between layers. As shown in the LFE results in Figure S1, the redshifts of peak C shows a very similar trend as the one in the previous version of our paper. Since the K-point in this calculation is  $10 \times 10 \times 8$  (due to the demand of the computational cost), we can only guarantee that the result is qualitatively correct.

For quasi-two-dimensional system—graphite, it is reported<sup>1</sup> that the LFE has an effect on the absorption energy at about 11 eV with an electric field perpendicular to the graphene layer, and the influent becomes more important when increasing the interlayer distance. Oppositely, also verified in our calculation, with decreasing the interlayer distance, the LFE becomes less important. For the absorption energy at low energy range, the impact of the LEF can be ignored. Thus, in our calculation within the LFE originated from the PBE-GGA approximation, the absorption trend is much similar to the one obtained with density-functional perturbation theory as we obtained also in this work.

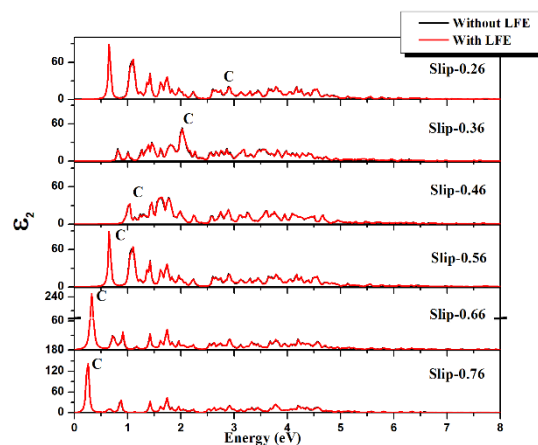


Figure S1. Imaginary part of the dielectric function of graphite under 100 GPa for a series of slip distances from AB-stacking to SP-stacking (0.36 Å, 0.46 Å, 0.56 Å, 0.66 Å, and 0.76 Å).with local field correction. It shows a similar trend with the results in our paper.

On the other hand, the neglect of long range interaction between graphene layers for compressed graphite can also be supported by our additional calculations for the same system but with the inclusion of VdW interaction. In the calculation, there is almost no change for our absorptions. Actually, it is reported<sup>2</sup> the VdW attractive interaction can be ignored when the interlayer distance is less than 3.0 angstrom. In our work, the obvious trend of the absorption is less than this distance. Thus, the VdW dispersion impact is little on our results. We have considered the VdW interaction with a relatively large interlayer distance (e.g. >40 GPa) and found almost no change of the optical absorption features. When interlayer decreases, the VdW interaction can be ignored, as pointed out in the previous paper<sup>2</sup>.

Typically, GGA always overestimates the bond length of the system due to its incomplete description in exchange-correlation interactions. Thus, the corresponding calculated

pressures in our paper are typically larger than the exact pressures. However, the trend of the absorption features is acceptable, even though the pressure evaluation may involve errors.

### The information of optimized structures of compression graphite

For optimized structures of graphite under compression along the crystallographic  $c$  direction, the graphene layers remain planar and show only a slight change in the intra-layer bond length. Even at 120 GPa, the bond along armchair direction (i.e.  $y$  direction denoted in Fig. 1(a)) is shortened by 0.015 Å, and for the other two bonds, one is elongated by 0.027 Å, while the other remains constant.

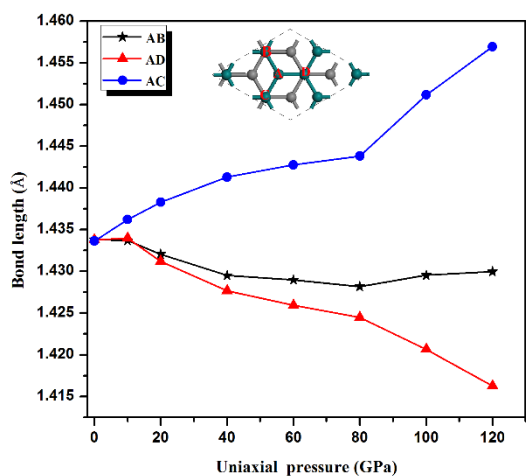


Figure S2 Bond length changes as a function of uniaxial pressure

### The definition of the orbitals mentioned in the manuscript

$\pi$  orbital is the hybridization of atomic  $p_z$  orbitals of the in-plane nearest carbon atoms, and  $\pi^*$  is the corresponding anti-bonding orbital.  $n$  orbital is defined as the atomic-like  $p_z$  orbital of carbon atoms.

### Band structures and wave functions of graphite without slip

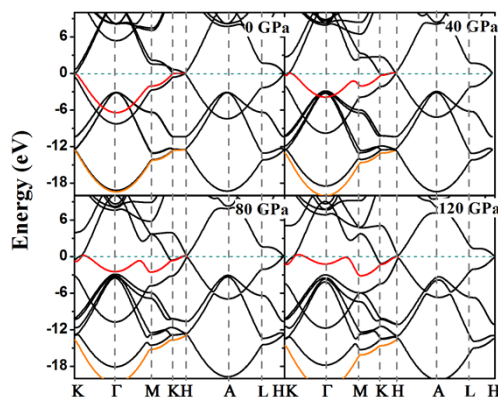


Figure S3 Band structure of AB-stacking graphite without slip under uniaxial pressures 0 GPa, 40 GPa, 80 GPa and 120 GPa. The red lines are the occupied  $\pi$  bands, which shift up towards the Fermi level with increasing pressure, while the dark yellow lines are the occupied  $\sigma$  bands, which split off and shift down with increasing pressure. The Fermi energy is set at zero.

### $\Delta\rho$ integrated over the $x$ - $y$ plane as a function of $z$ (along the $c$ axis)

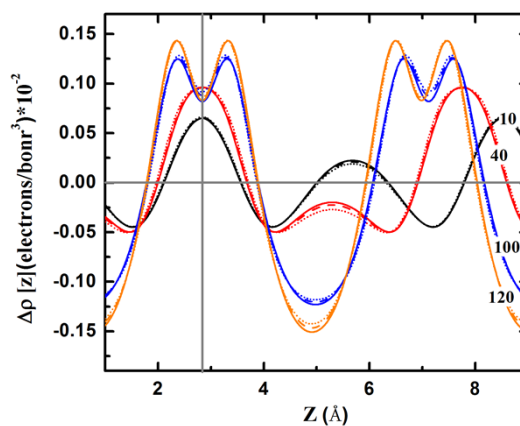


Figure S4  $\Delta\rho$  integrated over the  $x$ - $y$  plane as a function of  $z$  (along the  $c$  axis). The results are shown at 10 GPa (black curves), 40 GPa (red curves), 100 GPa (blue curves) and 120 GPa (yellow curves). The results with slip distance 0.0 Å, 0.36 Å and 0.70 Å are shown with solid lines, dashed lines and dotted lines, respectively.

### Supplemental references:

1. A. G. Marinopoulos, Lucia Reining, Angel Rubio, and Valerio Olevano, *Phys. Rev. B*, 2004, **69**, 245419.
2. M. Reguzzoni, A. Fasolino, E. Molinari, and M. C. Righi, *Phys. Rev. B*, 2012, **86**, 245434.