MS.7.P208
Interaction between electronic and mechanical degrees of freedom in graphene: probing by electron diffraction

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Keywords: graphene, corrugation, flexural phonons

Graphene can be considered as a specific form of matter – an electronic membrane [1,2,3]. So that the structure of a graphene sheet is formed in a strong interaction with charge carriers, which are easily generated in gapless graphene. It is well known that free-standing graphene undergoes severe corrugation [4]. The main reason for this is flexural phonons or, in other word, out-of-plane thermal oscillation which amplitude reaches several interatomic distances even at room temperature. The corrugation can also be induce by ad-atoms on graphene surface. Independently on its nature deformation of the crystal structure causes local shift of energy levels and, because of zero gap, considerable changes in charge carriers density distribution [5]. The strong coupling of the electronic behaviour with surface morphology makes graphene a sophisticated material to study.

Theory of the corrugation induced by thermal oscillations is quite non-trivial because of strongly anharmonical dynamics of flexural phonons [6]. Various models are presented in literature which differ significantly concerning the short wave-vectors range of the flexural phonon spectrum. Taking into account strain-induced charge carriers density inhomogeneities enormously complicates the task. For example, the effect suggests a specific thermal dependence of the graphene electronic properties; however, the problem is still unresolved [7].

In order to get some experimental knowledge on the issue of charge carriers-corrugation interaction we urged electron diffraction, which has shown itself as a useful tool for assessment of the corrugation of suspended graphene sheets [8]. A dynamic corrugation induce by flexural phonons can not be seen by direct imaging or low-speed scanning used by probe microscopies. At the same time, electron diffraction provides information about rapidly changing corrugation because of small time of the fast electron interaction with the specimen. Using electron diffraction one gets square of the corrugation spectrum averaged over the time of exposure, which can be extracted from diffraction tilt series. The feature of the diffraction imaging in transmission electron microscopes (TEM) is the high spatial coherency of the electron beam, which reaches 50nm in case microscopes equipped with field-emission electron gun. It allows to reach wave-vectors as small as 10^{-2}Å^{-1} in our measurements of the corrugation spectrum. From the right side the range is limited by low amplitude of the spectrum, which decrease as q^{-4} at this range, and can be estimated as 0.5 Å^{-1}.

. Interaction of the electron beam, used for obtaining of diffraction patterns, induces generation of electron-hole pairs in graphene, thus, increasing charge carriers density, which might have impact on corrugation state. One can vary charge carriers density by changing intensity of the beam. Measuring corrugation spectrum at different beam intensities provides an experimental evidence of the influence of the graphene electronic state on its structure. We have found shift of the harmonic-anharmonic border in flexural phonon spectrum with increasing beam current density, which designates the influence of increased charge carriers density. Change in small wave-vector range of the spectrum were found to be more prominent, since average electron wave-vector lays in this range.

It is very important to exclude possible effect of temperature change on our measurements, since electron beam also causes a Joule-heating of the specimen. In order to do this, we performed measurements of the corrugation spectrum under constant heating at temperatures up to 200ºC and found some change slight change in long wave-vector range of the spectrum. Our estimations of the sheet temperature increase due to effect of the incident electron beam show, that taking into account the superior thermal conductivity of graphene it is unlikely to be higher than several centigrade. So that the effect of the specimen temperature change in our experiments with varying beam currant density might not cause serious errors.

Thus, influence of the charge carriers density on corrugation of suspended graphene can be measured and degree of the specific electron-phonon coupling estimated.

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