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PHONONS IN BILAYER GRAPHENE

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Phonon properties of bilayer graphene have been investigated within Born — von Karman type lattice dynamics approach. A very good agreement between calculated phonon energy spectra of bilayer graphene and experimental data of bulk graphite has been obtained. Zone-center acoustic and optical vibrational modes were also analyzed. Obtained theoretical results can provide significant information on layer number and stacking configuration of graphene multilayers.

Keywords: phonons, graphene, lattice dynamics, Born — von Karman model.

Graphene attracts significant attention due to its unique electrical [1], mechanical [2] and thermal [3] properties. Graphene layers are considered as promising candidates for material base of future nanoelectronics [4]. Phonon (vibrational) properties of graphene are also of fundamental scientific interest. Phonon spectrum determines a series of important physical quantities, e.g. sound velocities, phonon density of states or thermal conductivity.

In present work, in the framework of the lattice dynamics approach, we investigate phonon properties of bilayer graphene with Bernal atomic stacking. Schematic view of considered structure is presented in Fig.1.

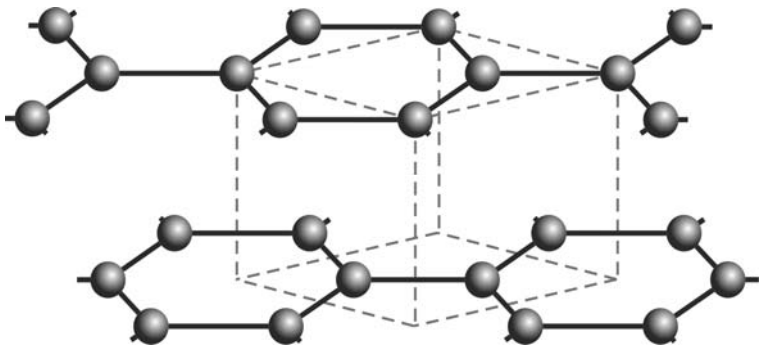

Fig. 1. Schematic view of Bernal bilayer graphene

For investigation of phonon properties of bilayer graphene we applied the Born – von Karman (BvK) model of lattice dynamics. Atomic vibrations were modeled taking into account interaction between atoms from four nearest-neighbor in-plane coordination spheres and from two out-of-plane spheres. Theoretical phonon dispersions were obtained by fitting the interatomic force constants of BvK model to experimental phonon frequencies of bulk graphite. In Fig. 2 we present the phonon energy spectrum for bilayer graphene, obtained within BvK model of lattice dynamics.

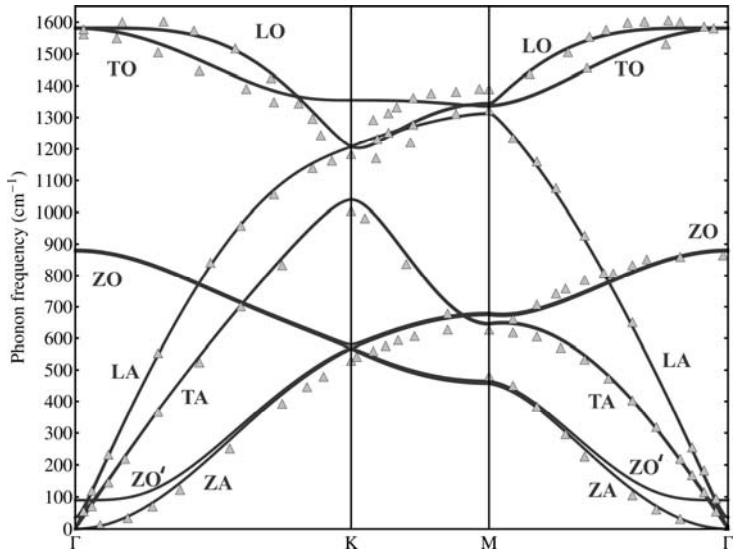

Fig. 2. Phonon energy spectrum of bilayer graphene. Experimental data points (gray triangles) for graphite from [5, 6] are shown for comparison

As can be seen from the figure, our calculated results are in a very good agreement with experimental data from [5, 6] for all phonon branches: in-plane acoustic branches (LA and TA), in-plane optic branches (LO and TO), out-of-plane acoustic (ZA) and optic (ZO) branches. A weak interlayer coupling between graphene layers (interlayer force constants are much less than intralayer ones) results in appearance of almost doubly degenerate phonon branches in the case of bilayer graphene compared with monolayer. The only exception constitutes the low-frequency ZO' mode with frequency of about 90 cm^{-1} at Γ point, which arises from an interlayer movement (see Fig. 3). An interesting behavior demonstrates out-of-plane acoustic ZA branch, in contrast to the linear dispersion near the Γ point for the in-plane TA and LA branches, it shows a q^2 dispersion, which is a characteristic feature for layered crystals [7, 8].

In Fig. 3 we show the atomic displacements in bilayer graphene, which correspond to different types of acoustic vibrations at Γ point.

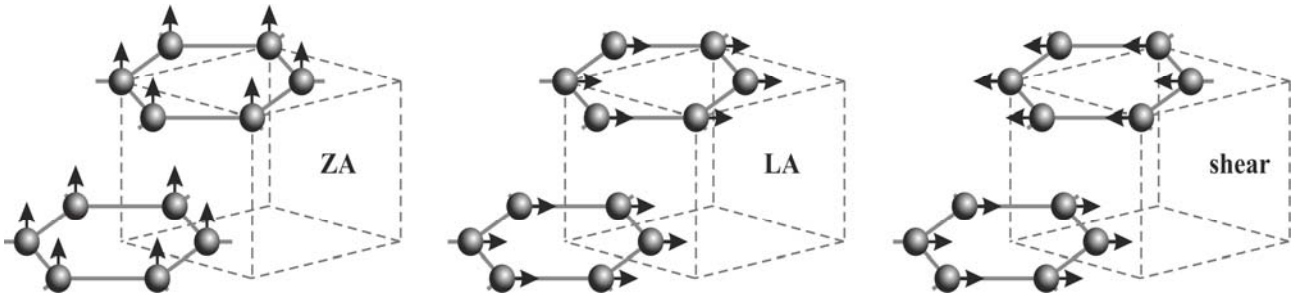

Fig. 3. Acoustic vibrations in Bernal bilayer graphene

In single-layer graphene there are only three types of acoustic modes at Γ point: out-of-plane (ZA), longitudinal in-plane (LA) and transversal in-plane (TA). As number of graphene layers increases from 1 to 2, an additional vibrational mode appears — in-plane shear, in which parallel graphene layers slide one over another in an opposite direction. This acoustic-like shear mode possessas non-zero frequency of about 36 cm^{-1} at the zone center and it has E_g symmetry type, which makes it Raman active and thus can be observed experimentally.

In Fig. 4 we show the schematic atomic displacements of zone-center optical vibrations in bilayer graphene.

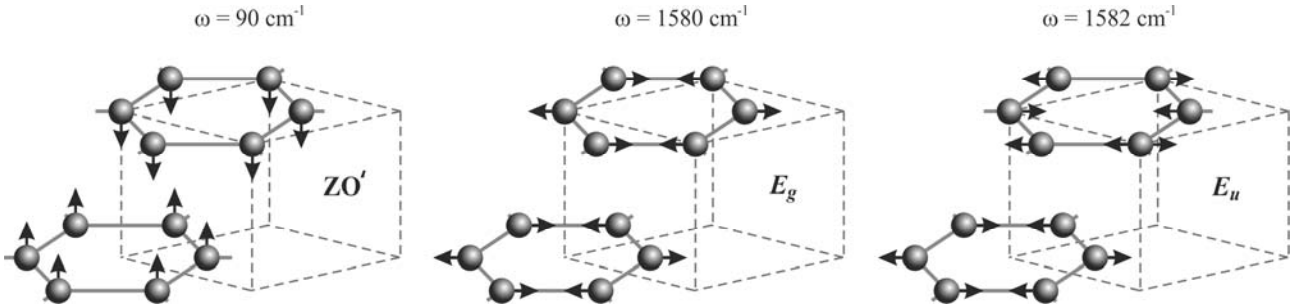

Fig. 4. Optical vibrations in Bernal bilayer graphene

In monolayer graphene exist three types of optical vibrational motion at Γ point: out-of-plane (ZO), longitudinal in-plane (LO) and transversal in-plane (TO). In bilayer graphene there are four atoms (in comparison with two atoms for single layer graphene) in the unit cell, therefore the zone-center optical vibration is more complex. In Fig. 4 few types of optical eigenmodes with different frequencies are presented. It is clearly seen from the figure that two splitted high frequency optical modes E_g and E_u are characterized by the in-plane vibrations, while the low-frequency ZO' mode arises from the interlayer motion along the *Z*-axis. Since in ZO' mode the top and bottom layers move in an opposite direction, it results in compression of graphene bilayer.

In conclusion, we applied a Born — von Karman model of lattice dynamics for investigation of phonon properties of Bernal bilayer graphene. In the framework of the developed theoretical approach we calculated phonon energy spectra of bilayer graphene in all high-symmetry crystallographic directions and obtained a very good agreement with experimental data for bulk graphite. We also analyzed zone-center acoustic and optical vibrational modes of bilayer graphene. Since many of them are Raman or infrared active, observation of these modes in experiments can be useful for analysis of layer number and stacking configuration of graphene multilayers.

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Фононы в двухслойном графене.

Исследованы фононные свойства двухслойного графена в рамках теории динамики решетки Борна — фон Кармана. Достигнуто отличное согласование между рассчитанными фононными энергетическими спектрами двухслойного графена и экспериментальными спектрами объемного графита. Также проанализированы акустические и оптические колебательные моды центра зоны Бриллюэна. Полученные теоретические результаты могут быть применены для анализа числа слоев и атомной конфигурации многослойного графена.

Ключевые слова: *фононы, графен, динамика решетки, модель Борна — фон Кармана.*