Phonons in ultrathin Bi(111) films: Role of spin-orbit coupling in electron-phonon interaction

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The vibrational properties of ultrathin Bi(111) films have been calculated by applying density functional perturbation theory with and without the inclusion of spin-orbit interaction. Phonon dispersion curves of dynamically stable bilayers are presented and analyzed. Surprisingly, the Rayleigh mode is found to have the largest amplitude on the second atomic layer. Moreover an optical surface phonon branch is found to be localized above the optical band and confined within the first surface bilayer. The analysis of this mode as a function of the number of bilayers allows one to estimate the contribution of the spin-orbit coupling to the electron-phonon interactions.

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I. INTRODUCTION

As one of the building blocks in topological insulator materials, such as Bi₂Se₃ and Bi₂Te₃, bismuth has attracted considerable interest. On its own bulk bismuth exhibits interesting properties. The bulk electronic band structure displays shallow electron and hole pockets¹ at high symmetry points in the Brillouin zone that give rise to a low carrier density of the order of 10¹⁷ cm⁻³.

Bismuth crystallizes in the A7 structure (R $\bar{3}m$ space group) and can be described either as rhombohedral with two atoms per unit cell, or as hexagonal with six atoms per unit cell (Fig. 1). Atoms are arranged in atomic bilayers perpendicular to the *c* axis according to the stacking AB-CA-BC, with stronger partially covalent bonds inside each bilayer and weaker bonds between adjacent bilayers. It may sometimes be convenient to view the structure as an fcc lattice distorted by a Peierls instability which causes a layer pairing along the $\langle 111 \rangle$ direction. Thus Bi cleaves more easily along $\langle 111 \rangle$ planes by breaking the weaker interbilayer bonds. Although a single bilayer is stable and found to be an insulator, the bulk originated from a sequential addition of bilayers evolves into a semimetal, whereas the surface acquires a metallic character.²⁻⁵ This is due to the emergence of surface electronic bands at the Fermi level which are spin split due to the large spin-orbit (SO) interactions, so as to form distinct hole and electron pockets along the $\bar{\Gamma} - \bar{M}$ direction of the surface Brillouin zone (SBZ), as revealed by angle resolved photoemission measurements (ARPES) and first-principles calculations.⁴⁻⁶ Such an important change of the electronic structure occurring at the surface is held responsible for Bi surface (interface) superconductivity, whereas no superconductivity has been found in bulk Bi.^{7,8}

These surface properties of Bi(111) largely justify a theoretical study of the phonon spectrum for ultrathin films. Indeed thin film physical properties change even more when the size of the system at hand is scaled down to, e.g., membranes, nanowires, and clusters. Ultrathin films of Bi(111) can be grown epitaxially^{9,10} on substrates such as Si(111)-7 $\times$ 7 where the interaction between the film and substrate is very

weak allowing for experimental studies of different physical phenomena without the influence of the substrate, as if the films were free-standing. The stability of the (111) bilayer structure—a feature common to the other group-V companions Sb(111) and As(111) and also black phosphorus—favors ultrathin films with an even number of atomic layers so as to permit the saturation of all *p_z* dangling bonds.^{9,11,12} Films with an odd number of (111) atomic layers are unstable and rearrange during the growth process into bilayer stacks.⁹

It has been shown that the role of spin-orbit interaction in bulk and low dimensional structures of bismuth plays a significant role in any realistic description of their electronic properties.^{13,14} As an example,^{15,16} differences in the phonon spectra calculated with and without the inclusion of spin-orbit interaction show that this interaction has important effects on the optical phonon modes of bulk bismuth.

As for the surface, the structure and dynamical properties of Bi(111) have been measured by low-energy electron diffraction¹⁷ and by inelastic helium atom scattering^{18,19} (HAS) technique. The latter technique measurements^{18,19} have shown a large surface corrugation of the order of 10% of the surface lattice constant, which remains practically constant as the temperature is varied. Moreover, the authors reported two low energy phonon branches in the acoustic region, one with character of the Rayleigh wave, the other with a flat dispersion between 3 meV at $\bar{\Gamma}$ and 4 meV at the $\bar{M}$ point.

In this work we investigate the structural and dynamical properties of ultrathin films of Bi(111) ranging from a single bilayer up to five bilayers by considering calculations with and without the inclusion of spin-orbit interaction. The focus will be on characterization of phonon modes localized in the uppermost bilayers and their evolution as the number of bilayers is increased.

II. COMPUTATIONAL DETAILS AND STRUCTURE

The total energy calculations were performed within the framework of the density functional theory (DFT)

FIG. 1. (Color online) Rhombohedral A7 structure of bulk bismuth. Internal parameters are listed in Table I. The right panel illustrates a top view of the Bi(111) structure while the lower panel shows a side view of the structure. The interlayer distances correspond to experimental values.

employing ultrasoft pseudopotentials. The dynamical properties of the Bi(111) bilayers were calculated by applying density functional perturbation theory (DFPT) as implemented in the QUANTUM-ESPRESSO²⁰ package. The pseudopotentials were generated according to the Rappe-Rabe-Kaxiras-Joannopoulos recipe.^{21,22} Local density approximation (LDA) was used for the exchange and correlation energy functional. The calculations were performed with and without the inclusion of the SO interaction.

The electron wave functions were expanded in plane waves with an energy cutoff of 35 Ryd, whereas for the effective potential and the charge density an energy cutoff up to 480 Ryd was used. The integrations over the surface Brillouin zone were carried out over a $(14 \times 14 \times 1)$ Monkhorst-Pack division of the $\mathbf{k}$ points, and a finite temperature Gaussian smearing of 0.02 Ryd was set when bilayers displayed metallic character. All calculations were carried out for fully optimized internal structure and lattice parameters.

As seen in Fig. 1 the two atoms of the rhombohedral unit cell of bulk Bi are located one at the origin while the second atom is at a fractional distance along the trigonal axis. Experimentally the fractional position of the second atom is at 0.4681 along the axis. The optimized structural parameters of the bulk are presented in Table I and are compared to the experimental values. The lattice parameter and the rhombohedral unit cell angle of the bulk structure are in a good agreement with the experimental values for both SO and no-SO calculations.

The Bi(111) bilayer thin films were constructed from the optimized bulk-terminated structure. Within this paper we consider one to five Bi(111) bilayer systems that are constituted by slabs with a vacuum region along the surface normal. The separating vacuum is set to 17 Å. The atomic layers were allowed to relax along the normal direction until the resulting forces acting on individual atoms were less than 25 meV/Å. The resulting interlayer relaxations with respect to bulk-terminated interlayer spacing are presented in Table II.

TABLE I. Structural parameters of the rhombohedral bulk Bi optimized with and without SO interaction. Lattice parameter a_0 , rhombohedral unit cell angle α , interatomic distances AB and AC, fractional position AB/(AC+AB), interlayer distance within one Bi bilayer, distance between two adjacent Bi bilayers, and in-plane lattice parameter of the Bi(111) surface.

	Bulk Bi (SO)	Bulk Bi (no-SO)	Expt. ¹⁷
a_0 (Å)	4.7220	4.6866	4.7236
α	57.33°	57.30°	57.35°
AB (Å)	5.5424	5.5313	5.5225
AC (Å)	6.2517	6.1773	6.2742
AB/(AB + AC)	0.4699	0.4724	0.4681
$d_{\text{Bi-Bi}}^{\text{short}}$ (Å)	1.611	1.628	1.594
$d_{\text{Bi-Bi}}^{\text{long}}$ (Å)	2.320	2.274	2.347
$a_{\text{in-plane}}$ (Å)	4.53	4.49	4.54

In this table the negative (positive) sign indicates a contraction (expansion) of the distance d_{ij} , between layer i and j with respect to the bulk-terminated value. The values in square brackets correspond to calculations where SO interaction was not included. The relaxation of the different bilayers considered is very similar when SO and no-SO calculations are compared. For one bilayer the resulting relaxation of the two atomic planes is a contraction with respect to the bulk value for both SO and no-SO calculations. Similarly, when two bilayers are considered, the intrabilayer distance d_{12} experiences a contraction while the interbilayer distance d_{23} is expanded. In all cases (one to five bilayers), the first intrabilayer distance is contracted by an amount which oscillates for increasing thickness between 1.5% [1.30%] and 2.3% [2.20%] when the SO coupling is [is not] included (Table II). Similar oscillations are predicted for the first interbilayer distance, d_{23} , and the second intrabilayer distance d_{34} , all characterized, like d_{12} , by a fairly slow convergence to the bulk values. Such a slow and oscillatory convergence may be associated with the considerable change of the electronic structure with increasing thickness, by which the insulating single bilayer gradually transforms into a semi-metallic bulk with a surface metallic character.

The dynamical properties of the relaxed Bi(111) bilayer thin films are calculated by evaluating the dynamical matrices on a $6 \times 6 \times 1$ grid of phonon wave vectors in the SBZ. The dynamical matrices are interpolated by taking the Fourier transform to find the real-space interatomic force constants. The real-space interatomic force constants are then obtained by Fourier transforming the dynamical matrix calculated over a thicker grid of points in the reciprocal space obtained by interpolation over the original $6 \times 6 \times 1$ grid. In this way the phonon dispersion curves of slabs of one to five bilayers are calculated at any desired wave vector along the SBZ high symmetry directions ($\bar{\Gamma} - \bar{M} - \bar{K} - \bar{\Gamma}$).

III. DYNAMICS OF BISMUTH SLABS

The phonon dispersion curves calculated with and without SO interaction for one to five Bi(111) bilayers are presented

TABLE II. Interlayer relaxation Δd_{ij} of one to five Bi(111) bilayers resulting from calculations with SO. The resulting values in square brackets correspond to no-SO interaction calculations.

	1	2	3	4	5
$\Delta d_{12}(\%)$	-1.54 [-1.30]	-2.27 [-2.00]	-2.00 [-1.60]	-1.95 [-1.50]	-2.21 [-1.50]
$\Delta d_{23}(\%)$	-	+1.30 [+1.69]	+0.63 [+1.30]	+0.47 [+0.77]	-0.08 [+0.90]
$\Delta d_{34}(\%)$	-	-	-1.70 [-1.96]	-1.15 [-1.40]	-1.60 [-1.25]
$\Delta d_{45}(\%)$	-	-	-	-0.32 [+1.12]	-1.31 [-0.90]
$\Delta d_{56}(\%)$	-	-	-	-	-1.30 [-0.60]

in Fig. 2. In both cases the Bi(111) bilayers considered in this report are all dynamically stable. Phonon dispersion curves corresponding to one bilayer [Fig. 2(a)] are characterized by three acoustic modes and three optical modes. The phonon polarizations of one Bi(111) bilayer at small wave vectors are ordered for increasing energy as shear-vertical (SV), shear-horizontal (SH), and longitudinal (L) for the acoustic branches, and SH, L, and SV for the optical branches. Unlike the paradigmatic layered crystal, graphite, where the two inequivalent atoms are in plane, the optical (SH, L) pair is well below the SV branch. At larger wave vectors, $\mathbf{Q}$, the acoustic SH approaches SV along the $\bar{\Gamma} - \bar{M}$ direction with a quasidegeneracy at $\bar{M}$. In the $\bar{\Gamma} - \bar{K}$ direction there is no mirror symmetry with respect to the sagittal plane and therefore the two shear branches can exchange their polarizations, so that the degenerate (SH, L) pair at $\bar{K}$ is below SV in the acoustic region, and vice versa in the optical region.

The acoustic modes calculated with SO interaction are softer than those calculated without SO interaction, especially for the optical branches, where for the single bilayer a softening as large as ~ 3 meV is produced at $\bar{\Gamma}$. Moreover, SO coupling induces interesting modifications of the dispersion curves of the single bilayer. The lowest SO acoustic branch shows a pronounced dip at about one-half of $\bar{\Gamma} - \bar{K}$ direction which does not occur in no-SO branch.

In the optical region the SO phonon branches start from the $\bar{\Gamma}$ point with opposite slopes with respect to the corresponding no-SO branches, so that they exhibit relative maxima at about 1/3 of both $\bar{\Gamma} - \bar{M}$ and $\bar{\Gamma} - \bar{K}$ directions. Similarly, SO optical branches acquire an upward curvature at $\bar{\Gamma}$.

As the number of bilayers is increased [Figs. 2(b)–2(e)] additional phonon branches appear, six per bilayer, that allow us to follow the development of surface modes and resonances, as well as the formation of bulk bands.

It is noted that the upper acoustic band edge is shifted upwards in energy as the bilayer number is increased. Since the acoustic modes mostly depend on the interbilayer force constants and the upper acoustic branch is that of SV modes with the largest amplitudes in the inner atomic layers, the upward shift is attributed with the slight reduction of the interbilayer distance from the surface region to the bulk. For the same reason the energy of the Rayleigh wave (RW), which in the continuum limit ($\mathbf{Q} \rightarrow 0$) is peeled off from the lower acoustic edge and has sagittal (SV $\times$ L) polarization, slightly decreases from an increasing bilayer number due to its exponential decay away from the surface, which is faster for larger values of $\mathbf{Q}$. At large $\mathbf{Q}$, however, the largest RW

amplitude occurs on the second and not on the first atomic layer, as more explicitly shown below.

The optical modes, which mostly depend on the intrabilayer force constants and are strictly related to the bilayer bonding structure, exhibit a more interesting dependence on the bilayer number. In particular the optical pair (SH, L) evolves for increasing thickness [Figs. 2(b)–2(e)] into the surface-localized Lucas modes with the characteristic degeneracy at $\mathbf{Q} = 0$.

In order to acquire a better understanding of which modes are localized at the surface or are surface resonances we display in Fig. 3 the phonon density of states (Ph-DOS) for a five-bilayer film, SO included, projected onto the atomic planes of the first bilayer. The solid lines correspond to projection of the Ph-DOS onto the first atomic layer and the dashed lines to projection onto the second atomic layer. The Ph-DOS is shown in Fig. 3 at the $\bar{\Gamma}$ point (a), at 1/3 (b), and 2/3 (c), along the $\bar{\Gamma} - \bar{M}$ direction and at the $\bar{M}$ point (d).

At the $\bar{\Gamma}$ point, Fig. 3(a), an intense SV feature is found at 13.6 meV with almost equal amplitude on both atomic layers of the first bilayer. The mode is split off from the upper optical bulk band edge. At deeper layers this peak is quite weak, which indicates a localization of this mode within the first bilayer. Additional features in the SV polarized Ph-DOS are found in the lower energy part of the spectra, namely, at 1.9, 3.9, and 5.5 meV. The Ph-DOS corresponding to L polarized modes display a broad character in the optical part of the phonon branches with a doublet of peaks, one at 12.2 meV and another one at 11.8 meV with equal displacement amplitudes in both first and second atomic layers. A third peak is found at 11.0 meV with the same characteristics as the former modes. In the lower energy part of the Ph-DOS the peaks with L character are found at 1.5 and 2.7 meV. At the $\bar{\Gamma}$ point, L and SH modes form degenerate pairs as required by symmetry. Along the $\bar{\Gamma} - \bar{M}$ direction [Figs. 3(b)–3(d)] the peak of the optical SV mode in the topmost bilayer shifts towards lower energy, whereas the L doublet centered at 12 meV shrinks into a single peak with practically no dispersion up to the $\bar{M}$ point.

To better understand the character of the modes and their components in the first and second atomic layer (first bilayer), as well as in the third atomic layer (second bilayer), the projected Ph-DOS's for the different atomic layers and polarizations can be conveniently represented by contour plots along the respective dispersion curves. In Fig. 4 we show the Ph-DOS's calculated for the five-bilayer slab with SO coupling included. The most noticeable feature in the Ph-DOS plots is the horizontal optical modes (SH and L modes) that exhibit a very flat dispersion along the SBZ directions and

FIG. 2. (Color online) Phonon dispersion curves along the SBZ high symmetry lines of Bi(111) bilayers from DFPT calculation with (solid lines) and without (dashed lines) SO coupling. Dispersion curves for (a) one bilayer, (b) two bilayers, (c) three bilayers, (d) four bilayers, and (e) five bilayers, respectively.

seemingly equal amplitude in the second and in the first atomic layer, respectively. The optical mode corresponding to SV displacements in the first atomic layer has instead a

FIG. 3. (Color online) Phonon density of states of five-Bi(111) bilayer film with SO included projected onto first (solid line) and second (dashed line) atomic plane resolved for shear-vertical (black) and longitudinal (red) polarizations plotted for different wave vectors, (a) at $\bar{\Gamma}$ point, (b) at $1/3$, (c) at $2/3$, and (d) at the $\bar{M}$ point along the $\bar{\Gamma} - \bar{M}$ direction. The arrows indicate the peak position of the highest optical mode in the absence of SO coupling.

downward dispersion towards the boundary of the SBZ. The Ph-DOS corresponding to the second layer optical transverse modes (SV_2 and L_2) displays a very distinct hybridization and an avoided crossing, where L_2 and SV_2 polarizations are exchanged in the vicinity of the $\bar{M}$ point. The conversion of the optical SV mode at $\bar{\Gamma}$ into an L mode at the zone boundary is quite evident in the third layer (SV_3 vs L_3).

In the acoustic region the lowest surface phonon branch, known in the long-wave limit as Rayleigh wave, is normally found to be localized below the bulk continuum with an elliptical polarization and the largest displacement in the first atomic layer. For ultrathin films of Bi(111) the lowest acoustic mode is surprisingly found to have the largest amplitude on the second atomic layer, whereas the SV acoustic mode localized on the first layer occurs at larger energies, with the largest amplitude at the $\bar{K}$ point. Since in an isolated bilayer the SV_1 and SV_2 modes form degenerate pairs by symmetry, the fact that in the five-bilayer sample the acoustic branch SV_2 is softer than SV_1 indicates that the interbilayer charge causes a softening of the second layer motion that the first atomic layer, facing vacuum, does not have.

IV. SPIN-ORBIT DEPENDENT ELECTRON-PHONON COUPLING

As seen in Fig. 5 the band structure near the Fermi level for no-SO (a) is quite different from that with SO interaction included (b), and also the evolution for increasing thickness from one (1BL) to five (5BL) bilayers is rather different in the two cases. These differences are clearly reflected in the way the energy of the optical SV_1 ($= SV_2$) phonon localized in the surface bilayer depends on the thickness with (d) and without

FIG. 4. (Color online) Contour plot of the phonon density of states for a five-bilayer film with SO included projected onto the first, second, and third atomic layer and onto shear-horizontal, longitudinal, and shear-vertical components of the mode polarization, respectively. The color scale of the intensities is ranged from yellow (lowest), green, cyan, blue to purple (highest).

(c) SO coupling, and we are able to extract some information on the SO-dependent electron-phonon interaction.

With no SO interaction [Fig. 5(a)] the 1BL band structure is that of an insulator with a gap of about 0.5 eV, but evolves into a semimetal already at 2BL, however, with a single electron pocket at $\bar{\Gamma}$ but a large gap at $\bar{M}$ and at $\bar{K}$, too. At a larger thickness small hole pockets arise around $\bar{\Gamma}$ with a reduction of the gap at the $\bar{M}$ point. With SO coupling included [Fig. 5(b)], a single bilayer is still an insulator, with a gap, however, of only 63 meV and the evolution towards a metallic character of the surface bilayer for increasing thickness is mostly due to the growth of broad hole and electron pockets in the $\bar{\Gamma} - \bar{M}$ direction and a gradual decrease of the gap in this direction, in agreement with ARPES data.^{1,14,23}

The corresponding changes in the optical SV_1 phonon energy at the symmetry points $\bar{\Gamma}$, $\bar{M}$, and $\bar{K}$ without and with SO coupling are shown in Figs. 5(c) and 5(d), respectively. Clearly the large change in the no-SO electronic structure from 1BL to 2BL (closure of the 0.5 eV gap) can be associated with the comparatively large (1.4 meV) drop of the $\bar{\Gamma}$ phonon energy. From 2BL to 5BL the electronic structure shows a fairly regular increase of the DOS at the Fermi level, which reflects a further slow decrease of the optical SV_1 phonon energy. Also the SV_1 phonon at $\bar{M}$ has a slow decrease with increasing thickness, which can be associated with the reduction of the electronic gap in the $\bar{\Gamma} - \bar{M}$ direction. On the contrary no substantial change in the electronic structure occurs in the $\bar{\Gamma} - \bar{K}$ direction, also consistent with the negligible change of the SV_1 phonon energy at $\bar{K}$ from 1BL to 5BL.

The band structure with SO coupling shows for increasing thickness the gradual formation at the Fermi level of the hole pocket along $\bar{\Gamma} - \bar{M}$ and the electron pocket ring around $\bar{\Gamma}$ [1] with a modest change of the DOS at the Fermi level. Accordingly the energy of the SV_1 phonon is practically constant for 1BL $\rightarrow$ 5BL at all three symmetry points of the SBZ, except a small softening at $\bar{M}$ for 1BL $\rightarrow$ 2BL. The important effect of SO interaction on the phonon energies is the overall softening which for 5BL and 1BL the optical SV_1 phonon at $\bar{\Gamma}$ amounts to $\Delta_{SO} = -1.5$ meV and $\Delta_{SO} = -3.0$ meV [Fig. 5(d)], respectively.

In DFPT the squared frequency of the surface phonon of wave vector $\mathbf{Q}$ and branch index ν (here $\nu = SV_1$) can be written as²⁴

$$\omega_{\nu}^2(\mathbf{Q}) = \omega_{\nu,0}^2(\mathbf{Q}) + \frac{1}{M} \sum_{l,l'} \xi_l^*(\mathbf{Q}, \nu) \left[\int d^3r n(\mathbf{r}) \nabla_l \nabla_{l'} V(\mathbf{r}) + \iint d^3r d^3r' \nabla_l V(\mathbf{r}) \chi(\mathbf{r}, \mathbf{r}') \nabla_{l'} V(\mathbf{r}') \right] \xi_{l'}(\mathbf{Q}, \nu),$$

where M is the Bi atom mass, $\xi_l(\mathbf{Q}, \nu)$ the phonon eigenvector, $n(\mathbf{r})$ the electron density, $\nabla_l \equiv \partial/\partial \mathbf{r}_l$ with l labeling the atoms at positions $\mathbf{r}_l$; $\omega_{\nu,0}^2(\mathbf{Q})$ is the squared frequency for bare (unscreened) ion-ion interaction, whereas the terms in squared parentheses express the effects of the interposed electrons subject to the total ion potential $V(\mathbf{r})$. This potential includes the Coulomb ion potential and the SO term, $V(\mathbf{r}) = V_{\text{ion}}(\mathbf{r}) + V_{\text{SO}}(\mathbf{r})$ and can be written as a sum over individual ion potentials, $V(\mathbf{r}) = \sum_i v(\mathbf{r} - \mathbf{r}_i)$. In this case the first term

FIG. 5. (Color online) Electronic band structure (Fermi energy $E_F = 0$) of one to five Bi(111) bilayers calculated without (a) and with (b) SO interactions. The evolution of the optical SV_1 phonon mode as a function of the number of bilayers calculated without (c) and with (d) SO interactions.

in the squared parentheses is diagonal with respect to (l, l') and for the SV_1 mode it gives approximately the contribution $(2M)^{-1} \int d^3r n(\mathbf{r}) [\partial^2 v(\mathbf{r} - \mathbf{r}_1) / \partial z^2 + \partial^2 v(\mathbf{r} - \mathbf{r}_2) / \partial z^2]$, where $\mathbf{r}_1$ and $\mathbf{r}_2$ are any atom position in the first and second layer, respectively. The second term, containing the electronic response function $\chi(\mathbf{r}, \mathbf{r}')$ expresses the effect of (nondiagonal) electron-phonon interaction, and is generally negative, yielding a phonon softening and, in case of $\chi(\mathbf{r}, \mathbf{r}')$ singularities, Kohn anomalies. It is directly proportional to the mode-selected electron-phonon coupling strength $\lambda_{\mathbf{Q}, \nu}$ ²⁵ and to the inelastic Helium atom scattering intensity from the $(\mathbf{Q}, \nu)$ th phonon.²⁶

The SO coupling acts on the vertex terms $\nabla_l V(\mathbf{r})$ and $\nabla_{l'} V(\mathbf{r})$, as well as on the response function $\chi(\mathbf{r}, \mathbf{r}')$ through the SO effects on the band structure around the Fermi level. The analysis of the results reported in Fig. 5 suggests that the SO coupling mainly operates through the vertex terms rather than through the response function (electron-hole propagator). One can note, e.g., that, despite the similarity between the

band structure near the Fermi level of no-SO 5BL and SO 3BL slabs [Figs. 5(a) and 5(b)], the average SO-induced softening remains of the order of -1 meV. From these considerations, albeit qualitative, it may be concluded that the SO part of the electron-phonon interaction associated with the vertex terms, more than the SO-induced change in the band structure, is responsible for the softening Δ_{SO} found in DFPT calculations with the SO-coupling included.

V. CONCLUSIONS

Our density functional perturbation theory calculations predict dynamical stability of ultrathin Bi(111) films. The inclusion of SO interaction turns out to be a rather crucial ingredient for the description of the higher energy optical part of the phonon branches while the acoustic modes are less influenced when compared with the calculations where SO interaction is not included. The optical phonons belonging to the higher energy part of the phonon spectrum display a rather flat dispersion within the SBZ, while some modes confined within the second atomic layer undergo hybridization that gives rise to avoided crossing in the vicinity of the $\bar{M}$ point where the mode acquires circular polarization.

The acoustic Rayleigh mode is found to belong to the second atomic layer rather than to the first one, as in ordinary clean nonlayered materials. The mode with a large SV_1 amplitude within the first atomic layer has a much higher energy and propagates within the continuum of phonon branches that belong to deeper atomic layers.

The analysis of the localized optical SV_1 mode, which is entirely confined in the surface bilayer for any thickness, as a function of the BL number for no-SO and SO calculations, indicates that the SO-induced softening can be attributed to the SO-dependent part of the electron-phonon interaction rather than to the SO-induced change of band structure. Actually the softening Δ_{SO} can be taken as an estimation of the SO-dependent part of the electron-phonon interaction.

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