I. INTRODUCTION

The discovery of graphene [1] opened a new and increasing research area on two-dimensional (2D) materials related both to fundamental issues and to many potential applications [2,3]. Among layered materials (LMs) [4–10], the transition-metal dichalcogenide (TMD) MoS2 has attracted attention due to its remarkable physical properties, such as high mobility (μ > 200 cm2 V−1 s−1) [11] and Young’s modulus (0.33 TPa) [12]. Besides, differently from graphene, MoS2 presents an intrinsic direct band gap of 1.8 eV [13], in the limit of monolayer thickness, which makes it a good candidate to complement (or even substitute) graphene in electronic [11,14] and optoelectronic [15–17] applications.

The bulk MoS2 single crystal is a semiconductor with a 1.3 eV indirect band gap [18]. It is built up from the stacking of S-Mo-S (monolayer) layers by weak van der Waals forces in a Bernal arrangement [19]. Each monolayer is formed by two hexagonal planes of sulfur (S) atoms sandwiching a hexagonal plane of molybdenum (Mo) atoms linked via ionic-covalent bonds arranged in a trigonal prismatic structure [18,20]. In the bulk form it belongs to space group P3m1 (point group D3h), whereas in the monolayer limit it belongs to P6m2 (point group D3h), and as a consequence monolayer MoS2 does not present an inversion symmetry [21]. This lack of inversion of symmetry splits the valence bands due to the spin-orbit interaction, opening possibilities for MoS2 as a suitable material for valleytronics [22]. The electronic gap of MoS2 can in fact be tuned by the number of layers [18].

The stacking effect on the E1g and A1g vibrational modes of mechanically exfoliated MoS2 samples supported on SiO2 was investigated by nonresonant Raman spectroscopy at high-pressure conditions. The split of E1g and A1g modes were observed for single- and bilayered samples whereas such splitting disappears for higher numbers of layers. The differences on the E1g pressure coefficients allowed us to interpret the observed splitting as due to the presence of two types of regions corresponding to a high and a low conformation of MoS2 to the substrate roughness. The difference in the pressure coefficient appears then as due to the biaxial stress introduced via the substrate compression. Such effects were not observed for the A1g mode due to its vibration symmetry. This out-of-plane vibration is mainly affected by the normal stress that corresponds to pressure transmitted by the pressure-transmitting medium.

Pressure application allows us to continuously increase the interaction between the 2D system and its environment, thus constituting a method of choice to understand such effects [23]. In this work the evolution of phonon modes with pressure will provide evidence that such interaction can be extremely inhomogeneous and can even have a bimodal character.

Raman spectroscopy is a nondestructive tool widely used to identify the numbers of layers of MoS2 thanks to its sensitivity to the MoS2 thickness variation [24,25]. Bulk MoS2 presents four first-order Raman-active vibrational modes in the center of the Brillouin zone, E2g, E1g, E1g, and A1g, located at about 32 cm−1, 286 cm−1, 383 cm−1, and 408 cm−1, respectively [20,26]. With the exception of E2g, all modes correspond to intralayer vibrations in a S-Mo-S layer, while the interlayer mode E1g comes from the respective atomic movement of two adjacent S-Mo-S layers to each other. The E1g mode is forbidden in a backscattering geometry perpendicular to the basal plane. The E1g mode results from the in-plane vibration of the sulfur and molybdenum atoms in opposite directions, whereas the A1g mode is related to out-of-plane vibrations of the sulfur atoms [20,25,26]. Because of experimental limitations, we investigated here only the behavior of E1g and A1g modes in n-layered MoS2.

The use of pressure has been an effective method to probe structural [7,27–30], electrical [27,31–33], and optical [13,34–36] properties of MoS2. It is known that the single crystal (as well as powder) of MoS2 experiences an electronic and structural phase transition (2Hc to 2Ha) under high pressure (∼20 GPa) governed by the sliding of adjacent S-Mo-S layers, leading to a semiconductor-metal transition when the phase change is completed [27,33]. For bilayer MoS2, high-pressure photoluminescence measurements were used to clarify the optical transitions that were not well established [13]. In a
FIG. 1. Identification of the MoS$_2$ thickness by comparing the frequencies of in-plane $E_{2g}^1$ (red squares) and out-of-plane $A_{1g}$ (red circles) vibrational modes of our sample (open symbols) with the results from Ref. [24], as well as their frequency differences $\omega_{A_{1g}} - \omega_{E_{2g}^1}$. The areas limited by the red dashed lines show the regions in which the Raman spectra were taken and the numbers correspond their number of layers. (b) Sketch of the atomic displacements of $E_{2g}^1$ and $A_{1g}$ modes.

recent report, high-pressure Raman measurements were used to investigate monolayer MoS$_2$. There, a pressure-induced phase transition characterized by the appearance of two new peaks located at 200 cm$^{-1}$ and 240 cm$^{-1}$ was observed at 5.8 GPa. This phase was suggested to be similar to the 17$'$-MoS$_2$ one, in which the sliding of S atoms inside the S-Mo-S layer is caused by a distortion of the unit cell [29].

Despite the availability of many studies about the influence of the stacking on the physical properties of MoS$_2$, the influence of stacking on the high-pressure response of MoS$_2$ remains less explored and constitutes an original method to study the interactions of LMs with their environment [37]. In this study we present the influence of the interlayer interactions due to thickness variation on the vibrational modes of the mono-, bi-, tri-, and many layers of MoS$_2$ submitted to hydrostatic high pressure by means of nonresonant Raman spectroscopy.

II. METHODOLOGY

Experimental setup and sample characterization

A few layers of MoS$_2$ were obtained by standard mechanical exfoliation [38] from a single crystal (SPI Supplies) and deposited on thin (50 $\mu$m thickness) cleaned silicon substrate with 300 nm of silicon oxide layer. The identification of the number of layers was done by optical contrast and Raman spectroscopy, Fig. 1. Raman spectra were acquired using the LabRAM HR800 from Horiba (during the pressure cycle) and Invia from Renishaw (ambient conditions) systems with energy excitation of 2.33 eV (532 nm) and power set at ~0.5 mW on the entrance of the high-pressure device to avoid thermal effects [39]. The laser was focused with a 50× magnification objective lens and the signal was dispersed by a grating of 1800 grooves/mm.

The Raman frequencies of in-plane $E_{2g}^1$ (open red squares) and out-of-plane $A_{1g}$ vibrational modes (open red circles) as a function of the number of layers is shown in Fig. 1. Also, their differences $\omega_{A_{1g}} - \omega_{E_{2g}^1}$ (open blue triangles) and the results from Ref. [24] (filled red and blue symbols) were placed for comparison. Our results show an excellent agreement with those found by Lee [24]. The Raman spectra were taken from the regions limited by red dashed lines on the inset of Fig. 1, which are labeled with their respective number of layers. Here the fits were done with a single Lorentzian peak to allow for comparison with the data of Ref. [24] in spite of the fact, as will be discussed later, that two components are present for each peak.

High-pressure experiments were performed using a diamond anvil cell pressure device with diamond anvils having a culet size of 700 $\mu$m. The MoS$_2$ sample consisting of regions with 1, 2, 3, and many layers deposited by mechanical exfoliation on a thin SiO$_2$/Si substrate was loaded in a cylindrical pressure chamber with diameter of ~315 $\mu$m and thickness of ~100 $\mu$m in a pre-indentated stainless steel gasket placed between the two diamond anvils. A 4:1 methanol-ethanol mixture, that is known to remain liquid and hydrostatic up to 10.5 GPa [40], was used as a pressure-transmitting medium (PTM). A small ruby chip was placed together with the sample and PTM inside the pressure chamber to calibrate the pressure by the standard ruby luminescence R1 line [41].

Spectra were collected for each region of the sample in the same hydrostatic conditions up to a maximum pressure of 8.7 GPa.

III. RESULTS

Figure 2 shows the Raman spectra of a monolayer (a), bilayer (b), trilayer (c), and many-layer (d) sample of MoS$_2$ ranging from ambient conditions (ACs) up to 8 GPa. We see that for the monolayer and the bilayer samples (at ambient conditions) the vibrational modes $E_{2g}^1$ and $A_{1g}$ (and their corresponding ones for odd number of layers, $E'$ and $A_1$, respectively) need to be fitted with two Lorentzian line shapes for each one. We have labeled these components as $E_{2g}^{(h)}$, $E_{2g}^{(l)}$, $E_{1g}^{(h)}$, $E_{1g}^{(l)}$, $A_{1g}^{(h)}$, and $A_{1g}^{(l)}$. The superscript can be for the moment considered as arbitrary, but corresponds to “in high conformation”($h$) and “in low conformation”($l$) as will be explained later. With increasing pressure, the intensity ratio between the ($h$) and the ($l$) components evolves. In particular, in the case of the monolayer sample the intensity of the ($h$) component becomes very weak after 0.7 GPa with...
the $E'(h)$ peak becoming undetectable beyond 1.6 GPa. The effect is less pronounced for the bilayer sample where the two components are always present at all the measured pressures.

For the trilayer and the sample having many layers (which can be assimilated to bulk MoS$_2$) we do not observe such splitting. The peaks in Figs. 2(c) and 2(d) are labeled $E_2^{(h)}$ ($E''$), $A_1^{(h)}$ ($A_1'$). We note in Figs. 2(b) and 2(c) that for the bilayer and trilayer samples, a new peak, which is labeled $B$, is visible from $\sim$5 GPa with its intensity increasing with pressure. Finally, in the bilayer sample a broad and weak intensity peak [labeled as $C$ in Fig. 2(b)] is also observed on the higher energy side of the $A_1'$ band appearing from 7.2 GPa.

To study the pressure dependence of the vibrational modes, we fit each mode with a Lorentzian function (Fig. 2) for all pressure points acquired and plot their Raman frequencies as a function of pressure as shown in Fig. 3. Here, all vibrational modes present linear and positive pressure coefficients, which are given in Table I together with the value of their frequencies at ambient pressure.
The obtained linear coefficient of the $E_{2g}^1$ mode of $1.7 \pm 0.1$ cm$^{-1}$/GPa$^{-1}$ for the many-layer sample is in rather good agreement with the values found in the literature for bulk MoS$_2$ which vary between 1.8 and 1.9 cm$^{-1}$/GPa$^{-1}$ [34,42,43]. The corresponding $A_{1g}$ value of the many-layer sample, $3.3 \pm 0.1$ cm$^{-1}$/GPa$^{-1}$, is on its side slightly smaller than the bulk values found in the literature which range from 3.6 to 4.0 cm$^{-1}$/GPa$^{-1}$ [34,42,43].

The obtained frequency of the $B$ mode and its pressure coefficient are close to the values observed in bulk MoS$_2$ for the $B_{1u}$ mode ($\omega_0 = 402.9$ cm$^{-1}$ and $\partial \omega / \partial P = 2.2$ cm$^{-1}$/GPa$^{-1}$) [34]. This mode is not Raman active and in fact it is not observed in all bulk experiments. The activation of this mode in the bilayer and trilayer sample at pressures about 5 GPa remains to be understood. The $C$ mode, only observed in the last two spectra of the bilayer sample, lies at $\sim 10$ cm$^{-1}$ above the $A_{1g}$ mode. This is also the case of dispersive modes observed in bulk MoS$_2$ interpreted as involving multiple phonon scattering with polariton coupling [34]. In the following our discussion will be centered on the $E_{2g}^1$ and $A_{1g}$ modes.

**IV. DISCUSSION**

Previous studies in mono- and bilayer MoS$_2$ deposited on flexible substrates submitted to uniaxial strain showed a splitting of the $E_{2g}^1$ modes which was interpreted as

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**FIG. 3.** Pressure dependence of MoS$_2$ Raman frequencies for all observed Raman vibrational modes and the corresponding linear fits (except for the $C$ mode in the bilayer which did not present a number of points enough to make a fitting reliable) for monolayer (a), bilayer (b), trilayer (c), and many-layer (d) samples.
TABLE I. Assignment of MoS$_2$ vibrational modes and their frequencies intercept ($\omega_0$), as well as their pressure coefficients ($\partial \omega_0 / \partial P$). The Raman modes labeled $B$ could not be unambiguously assigned and $n$ stands for the number of layers.

<table>
<thead>
<tr>
<th>$n$</th>
<th>$E^{(i)}_{2g}$ ($E^{(ii)}$)</th>
<th>$E^{(i)}_{2g}$ ($E^{(ii)}$)</th>
<th>$A^{(i)}_g$ ($A^{(ii)}_g$)</th>
<th>$A^{(i)}_g$ ($A^{(ii)}_g$)</th>
<th>$B$</th>
</tr>
</thead>
<tbody>
<tr>
<td>1</td>
<td>383.8</td>
<td>384.9</td>
<td>408.4</td>
<td>404.5</td>
<td>2</td>
</tr>
<tr>
<td>2</td>
<td>383.5</td>
<td>384.4</td>
<td>408.3</td>
<td>405.9</td>
<td>3</td>
</tr>
<tr>
<td>3</td>
<td>383.8</td>
<td>407.8</td>
<td>399.2</td>
<td></td>
<td>many</td>
</tr>
</tbody>
</table>

$\omega_0$ (cm$^{-1}$) | $\partial \omega_0 / \partial P$ (cm$^{-1}$/GPa)

1 | 2.0 $\pm$ 0.1 | 3.3 $\pm$ 0.4 | 3.6 $\pm$ 0.1 | 3.6 $\pm$ 0.1 |
2 | 2.1 $\pm$ 0.1 | 2.6 $\pm$ 0.1 | 3.5 $\pm$ 0.1 | 3.6 $\pm$ 0.1 |
3 | 1.9 $\pm$ 0.1 | 3.5 $\pm$ 0.1 | 3.0 $\pm$ 0.3 |
many | 1.7 $\pm$ 0.1 | 3.3 $\pm$ 0.1 |

A breaking of the degeneracy in the initially degenerate in-plane mode $E^{(i)}_{2g}$ [44,45]. Such breaking of the degeneracy was interpreted as due to van der Waals interactions at the substrate-sample interface. For bulk MoS$_2$ the in-plane modes $E^{(i)}_{2g}$ and $E^{(ii)}_{2g}$ also exhibit splitting at about 20 GPa caused by the modification in the interlayer stacking through the sliding between adjacent S-Mo-S layers [27,33,34]. Similar splits for both vibrational modes $E^{(i)}_{2g}$ and $A^{(i)}_g$ in monolayer MoS$_2$ under high pressure have been observed [29]. There, they were explained by the deformation of the S-Mo-S structure caused by different interactions of the silicon substrate and PTM with the S layers, which can induce sliding of Mo or S layers within the monolayer, leading to different Mo-S bond lengths and consequently to different vibration energies. Our measurements on monolayer MoS$_2$ and those of Ref. [29] are in excellent agreement. Nevertheless our observation of the presence of the same type of splitting of the $E^{(i)}_{2g}$ and $A^{(i)}_g$ modes also for the bilayer sample and its absence for a higher number of layers lead us to a different interpretation.

In the case of graphene, Nicolle et al. [37] observed a different response in the high-pressure Raman signal for mono- and bilayer in comparison with trilayer samples. Monolayer and bilayer graphene undergo a biaxial strain resulting from the substrate volume reduction on pressure application. The graphene ability to conform to the SiO$_2$ substrate roughness is the key point for such biaxial transmission. Unlike the mono- and bilayer samples, trilayer graphene undergoes a 3D hydrostatic compression by the PTM due to the unbinding state between SiO$_2$ substrate-trilayer graphene, related to the higher bending modulus of the 3-layered sample.

The parameters governing the conformations of a 2D system to a rough substrate are the adhesion energy ($\gamma_a$), the bending modulus of the 2D $n$-layer system ($D_n$), and the characteristic curvature radius of the surface ($k_r$). It has been shown [46] that in 2D membranes a single dimensionless parameter, $\alpha = (k_{es}/k_c)^{1/2}$, which compares the typical curvature of the substrate with the adhesion equilibrium curvatures, $k_{es} = (2\gamma_a/D_n)^{1/2}$, governs the conformation of the 2D membrane to the surface. Perfect adhesion is expected for $\alpha > 0.8-0.86$ [46] for graphene on SiO$_2$/Si and by using a characteristic $k_r = 0.3$ nm$^{-1}$, leads to an unbinding when the number of graphene layers is increased from 2 to 3 [37]. The substrates used in the present work are in fact the same used in Ref. [37]. In the case of single-layer MoS$_2$ its bending modulus, $D_1$, has been calculated to be 9.61 eV, i.e., ~7 times higher than the value for single-layered graphene [47]. On the other side, it has been shown in AFM experiments [48] that the adhesion energy of MoS$_2$ on SiO$_2$ is smaller than for graphene on SiO$_2$. The report between graphene and MoS$_2$ $D_1$ values being more important than the report between $D_1$ and $D_2$ in graphene (see later), we may expect that an unbinding transition of MoS$_2$ on SiO$_2$ can already take place in the monolayer system.

This is supported by AFM observations [49] in which the roughness ratio between single-layer MoS$_2$ on SiO$_2$ and bare SiO$_2$ was found to be 0.56 while the equivalent ratio reported for graphene is 0.9 or higher [50]. We then expect that for $n = 1$ MoS$_2$ can already be found in a low-conformation state. The membrane model here referred to [46] is based on the description of the surface roughness as a periodic fixed curvature structure. Real roughness leads to a statistical distribution of curvature values and we admit that MoS$_2$ can be found in alternating regions in high conformation with the SiO$_2$ substrate ($h$) and in low conformation with the substrate ($l$). The two components of both the $E^{(i)}_{2g}$ and $A^{(i)}_g$ modes in our sample can be explained by considering this particular situation. In the $h$ region the MoS$_2$ layer, in adhesion with the substrate, undergoes a biaxial strain due to the volume reduction of the substrate on pressure application. On the other hand, in the $l$ regions the PTM flows through unbound regions and then the MoS$_2$ layer experiences a reduced biaxial strain, with a response closer to hydrostatic.

In that way the two components appearing in the $E^{(i)}_{2g}$ and $A^{(i)}_g$ in mono- and bilayer MoS$_2$ can be well interpreted as due to the presence of both highly and lowly confined regions in each one of these samples. The differences in interaction with the surface explains the splitting in the energy position of these Raman modes. As can be seen in Table I, the pressure evolution of the out-of-plane modes [$A^{(i)}_g$ ($A^{(ii)}_g$), $A^{(ii)}_g$ ($A^{(ii)}_g$)] and of the unbound $E^{(i)}_{2g}$ ($E^{(ii)}_h$) mode are within error bars independent of the number of layers or at most a slightly decreasing function of $n$. On the other side, the $E^{(i)}_{2g}$ ($E^{(ii)}_h$) mode exhibits a very different pressure slope with respect to the $E^{(i)}_{2g}$ ($E^{(ii)}_h$) mode as well as a strong dependence on $n$ between the monolayer and the bilayer sample. We will come later to provide an explanation to this effect.

We note the absence in the trilayer sample as well as the many-layer one of any Raman component corresponding to regions highly conformed to the surface roughness. This can be easily understood from the evolution of the bending rigidity and adhesion with the number of layers. In fact, the bending rigidity $D_n$ is an increasing function of $n$ which varies as $D_n = D_1 n^3$ assuming that sliding between the layers is negligible. In the case of graphene, experiments [51] show an even faster evolution with $n$ than the above referred to power law: $D_{\text{graphene}}(1) = 7.1^{+4.0}_{-3.0}$ eV, $D_{\text{graphene}}(2) = 35.5^{+20.0}_{-15.0}$ eV, and $D_{\text{graphene}}(3) = 126^{+71.0}_{-53.0}$ eV. We should note that this $D_1$ value seems to be overestimated when compared with the different values compiled from the literature [52]. The adhesion energy, $\gamma_a$, on its side should be a weakly varying function
of \( n \). We then expect that the ratio of lowly confined and highly confined surfaces, \( S_1(n)/S_0(n) \), is a rapidly increasing function of \( n \). Our experiments show that in MoS\(_2\) lying on SiO\(_2\) substrate with as-grown roughness, \( S_0(n) \to 0 \) for \( n > 2 \).

The differences in the bending rigidity between the monolayer and the bilayer samples can also explain the observed differences between the high-pressure behavior of their high-conformation components. First we may consider the differences concerning the intensity of the signal. The rapid attenuation in the monolayer sample of the \((h)\) components of the Raman signal can be interpreted as due to a rapid increase of the value of \( \theta \) as the pressure is transmitted as a biaxial stress to the 2D sample if that layer is valid [54]. The substrate deformation by the effect of pressure is transferred from the PTM to sample. However, for 2D systems supported on a substrate this assumption is not valid [54]. The substrate deformation by the effect of pressure is transmitted as a biaxial stress to the 2D sample if that layer is well conformed to the substrate. Consequently, a plot of Raman shift versus pressure is not appropriate, since the pressure derivative does not represent an intrinsic response of the 2D material, but it depends on the mechanical properties of the substrate. We need then to consider the biaxial stress transmitted from the SiO\(_2\) substrate to the MoS\(_2\) layers. The MoS\(_2\) biaxial stress is linked to the applied pressure through [54]

\[
\sigma_{\text{MoS}_2} = \theta(n) \frac{\beta_{\text{MoS}_2}}{\beta_{\text{SiO}_2}} \frac{\partial \omega_{\text{E}^{1(\text{h})}}}{\partial P}, \tag{1}
\]

where \( \theta(n) \) is the strain transfer efficiency, that can vary from 1 (total strain transfer) to 0 (fully unbinding MoS\(_2\) from the SiO\(_2\)). This parameter is dependent on the number of layers and on the substrate properties [54]. In our case, the substrate is the same for all \( n \) values and we can then assume \( \theta(n) \) as being only dependent on the number of layers. The parameters \( \beta_{\text{MoS}_2} \) and \( \beta_{\text{SiO}_2} \) are the linear in plane stiffness constants of MoS\(_2\) and of the substrate, respectively, having values of \( \beta_{\text{MoS}_2} = 190 \text{ GPa} \) [28] and \( \beta_{\text{SiO}_2} = 114 \text{ GPa} \) [54]. This equation gives us a relation between the pressure slope of those Raman modes involving bond vibrations in the direction of the biaxial stress, i.e., in our case the \( E_{2g}^{1(\text{h})} \) mode, and the pressure coefficient of the corresponding Raman mode in the bulk:

\[
\frac{\partial \omega_{E_{2g}^{1(\text{h})}}}{\partial P} \approx \theta(n) \frac{\beta_{\text{MoS}_2}}{\beta_{\text{SiO}_2}} \frac{\partial \omega_{\text{E}^{1(\text{h})}}^{(\text{bulk})}}{\partial P}, \tag{2}
\]

where we have assumed as a zero-order approximation that for high values of \( \theta \) the in-plane Raman mode evolution is essentially governed by the biaxial strain effects. Considering the values in the literature for \( \frac{\partial \omega_{E_{2g}^{1(\text{h})}}^{(\text{bulk})}}{\partial P} \) ranging from 1.7 to 1.9 cm\(^{-1}\)/GPa\(^{-1}\) leads to a value of \( \frac{\partial \omega_{E_{2g}^{1(\text{h})}}}{\partial P} = 3.0 \pm 0.3 \text{ cm}^{-1}/\text{GPa}^{-1} \) when taking \( \theta(1) = 1 \). This value is in excellent agreement with the experimental result in single-layer highly confined MoS\(_2\), meaning that the fraction of the surface being highly conformed to the surface, \( S_0(1) \), is also highly biaxially strained \( \theta(1) \sim 1 \), explaining the higher pressure slope of the \( E^{(\text{b})} \) mode. In addition when expressed as function of the strain we obtain \( \frac{\partial \omega_{\text{E}^{(\text{b})}}}{\partial \epsilon} = 3.8 \pm 0.4 \text{ cm}^{-1}/\% \) in rather good agreement with published values [44].

In the case of \( n = 2 \) the fact that \( \frac{\partial \omega_{E_{2g}^{1(\text{h})}}^{(\text{bulk})}}{\partial P} < \frac{\partial \omega_{E_{2g}^{1(\text{h})}}}{\partial P} \) means that \( \theta(2) < \theta(1) \) and should tend to zero for \( n = 3 \). The determination of the exact value of \( \theta(n) \) for \( n > 1 \) needs a deeper understanding on the compression process and in particular on the simultaneous participation of hydrostatic and biaxial strain components on a system which contrarily to graphene involves out-of-plane chemical bonds. In Fig. 4 are shown the different values for the \( E_{2g}^{1(\text{h})} \) and \( A_{1g} \) pressure slopes highlighting the transition from the bimodal adhesion behavior for \( n = 1, 2 \) to single low-conformation adhesion for \( n > 2 \).

To conclude, it is interesting to consider the reasons that the mechanical response of MoS\(_2\) on SiO\(_2\) at high pressure differs significantly from the one of graphene on SiO\(_2\). We can underline two distinctive aspects:

1. MoS\(_2\) shows for \( n = 1, 2 \) the presence of mixed highly and lowly confined regions and total unbinding for \( n > 2 \). In graphene such bimodal behavior was not observed. Graphene shows strong adhesion for \( n = 1, 2 \) and total unbinding from a

![FIG. 4. Pressure slopes of the \( E_{2g}^{1(\text{h})} \) and \( A_{1g} \) modes as function of the number of MoS\(_2\) layers. The two different high [(h), black] and low [(l), red] conformation components are separated. The lines are guides for the eye. We have underlined the sensitivity of the \( E_{2g}^{1(\text{h})} \) component to the biaxial strain generated by substrate compression. The drawings at the bottom of the figure sketch the bimodal and single adhesion situations.](image-url)
SiO$_2$ substrate for $n > 2$ [37]. Such differences are related to the higher bending modulus of MoS$_2$.

(2) For single-layer MoS$_2$ on SiO$_2$, the highly conformed regions have values of $\theta(1) \sim 1$ whereas for graphene on SiO$_2$, the biaxial strain transfer is much smaller with $\theta(1) \sim 0.2$ [54]. Such strong difference is explained from the strong differences on in-plane compressibility between MoS$_2$ and graphene with respect to the one of the SiO$_2$ substrate. Graphene being much less compressible is not able to follow the deformations imposed by the high biaxial strain provided by the SiO$_2$ substrate during its compression, whereas the highly conformed MoS$_2$ regions, with an in-plane compressibility much closer to the one of silica, deforms at the same rate that the substrate.

**V. CONCLUSIONS**

We performed a high-pressure nonresonant Raman study on mechanically exfoliated MoS$_2$ deposited on SiO$_2$. A splitting of the $E_{2g}$ and $A_{1g}$ modes was observed for the $n = 1$ and 2 samples which is enhanced with increasing pressure. Such splitting, absent for $n > 2$, is interpreted as due to the simultaneous presence of regions highly and lowly conformed to the substrate roughness, respectively. Only the regions highly conformed to the surface show a significant evolution of the $E_{2g}$ mode pressure slope with the number of layers. The absolute values of such slopes are explained in terms of the biaxial strain transfer from the substrate and its evolution with the number of layers as due to the evolution of the efficiency of the strain transfer from the substrate. The differences of in-plane compressibility between MoS$_2$ and the substrate and the evolution of the bending rigidity of MoS$_2$ with the number of layers are discussed as being the physical parameters governing the observed behavior.

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