(001)-surface-induced bulk states and surface resonances in II-VI zinc-blende semiconductors

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In a previous paper [Phys. Rev. B 50, 1980 (1994)] we gave an account of the nondispersive band found experimentally at 4.4 eV for CdTe(001) by Niles and Höchst. We have characterized this band as a surface-induced bulk state. In a second paper we showed that a similar state does exist in II-VI and III-V zinc-blende semiconductor compounds. In this paper we show that there are more such states within the valence-band energy interval. We use tight-binding Hamiltonians and the surface-Green’s-function matching method to calculate the surface resonances and surface-induced bulk states in the zinc-blende semiconductors CdTe, CdSe, ZnTe, and ZnSe. We find one distinctive surface resonance for the cation- and two for the anion-terminated (001) surface and three (001)-surface-induced bulk states with energies that correspond to the value of the heavy-hole, light-hole, and spin-orbit bands at X.

I. INTRODUCTION

The study of the physics of surfaces, interfaces, quantum wells, and superlattices of semiconductors has been of interest in the last few years.1–9 The interest is not only in binary compounds but also, more recently, in ternary and quaternary compounds.

The starting point is an accurate description of the electronic band structure of the binary compound by a method that serves as a sound basis for a clear and simple description of the more complicated compounds and systems.

In previous work10,11 we have used the tight-binding formulation to calculate the local density of states (LDOS) and, in conjunction with the known surface-Green’s-function matching method (SGFM),12 to derive the surface and the interface LDOS. In this paper we want to use the same method to calculate the surface resonances and surface-induced bulk states. The method can also be applied to superlattices9,13 and phonons.14 These band structures can also be used to calculate transport properties in heterostructures such as quantum wells, for example, by making use of the well known many-body formulation by Keldysh.15

In this paper, we consider the band structure of the II-VI semiconductors CdTe, ZnTe, CdSe, and ZnSe. We have obtained the bulk bands (infinite medium) from the direct diagonalization of the tight-binding Hamiltonians and from the poles of the real part of the corresponding Green’s function. These very same results were obtained using the SGFM method, from the (001)-bulk-projected Green’s function. This is actually a proof of consistency, which gives us confidence in the results.

Our calculation shows the existence of three surface-induced bulk states, which have no dispersion from Γ to X and do not change in energy with different atom termination of the sample. These states were found experimentally by Niles and Höchst3 in CdTe(001). There is also a distinct surface resonance for each anion- and cation-terminated surface. They do differ noticeably in energy. A general pattern develops for the location of the different bands in II-VI zinc-blende semiconductors within the range of their valence-band energy.

The rest of the paper is organized as follows. In Sec. II, to make the paper self-contained, we briefly describe the main highlights of the method and present the formulas that we have used. In Sec. III, we introduce the general characteristics of the valence band and the location in energy of the surface resonances and surface-induced bulk states. Next, we discuss the results for each system studied. In a final section, we present our conclusions. We have included an Appendix where we quote all the tight-binding parameters of interest for this calculation.

II. METHOD

We make use of tight-binding Hamiltonians. Since the Green’s-function matching method takes into account the perturbation caused by the surface exactly, at least in principle, we can use the tight-binding parameters (TBP’s) for the bulk.10,11,16 This does not mean that we are using the same TBP for the surface and the bulk. Their difference is taken into account through the matching of the Green’s functions. We use the method in the form cast by García-Moliner and Velasco.12 They use the transfer matrix approach first introduced by Falicov and Yndurain.17 This approach became very useful due to the quickly converging algorithms of López-Sancho et al.18 Following the suggestions of these authors, the algorithms for all transfer matrices needed to deal with these systems can be found in a straightforward way.19 This method has been employed successfully for the description of surfaces,5,10,18 interfaces,11,20 and superlattices.9,13
The formalism

We have first calculated the bulk (infinite medium) band structure of the compounds by the tight-binding method in the Slater-Koster language\textsuperscript{21} using an orthogonal basis of five orbitals, \( sp^3s^* \). The \( s^* \) state is introduced to properly locate in energy the conduction band usually formed by \( d \) states in the II-VI zinc-blende semiconductor compounds.\textsuperscript{22,23} In our calculation, we have included the effect of the spin-orbit interaction.\textsuperscript{24} The TBP's that we have used in our calculation are listed in the Appendix. They reproduce the known bulk bands quite well.\textsuperscript{5,25,26} We assumed ideal truncation.

We obtain the Green's function from

\[
(\omega - H)G = I ,
\]

where \( \omega \) is the energy eigenvalue, \( H \) is the tight-binding Hamiltonian, and \( I \) is the unit matrix. We adopt the customary description in terms of principal layers. We label them with positive numbers and zero for the surface principal layer. Atomic layers are labeled with negative numbers and zero for the atomic surface layer. Let \( | n \rangle \) be the principal wave function describing the \( n \)th principal layer. It is a linear combination of atomic orbitals wave function with one \( s \)-like, three \( p \)-like, and one \( s^* \)-like atomic functions per spin on each atom in the unit cell.

(There are two different atoms in the cell, and two atomic layers per principal layer, i.e., it is a 20-dimensional vector.) If we take matrix elements of Eq. (1) in the Hilbert space generated by the complete set of the wave functions \( | n \rangle \), we get

\[
\langle n | (\omega - H)G | m \rangle = \delta_{nm}.
\]

Since there are only nearest-neighbors interactions between principal layers, the identity operator for the \( n \)th principal layer is

\[
I = | n - 1 \rangle \langle n - 1 | + | n \rangle \langle n | + | n + 1 \rangle \langle n + 1 |,
\]

and therefore \( H_{m,m+i} = 0 \) for \( i \geq 2 \). By inserting Eq. (3) into Eq. (2), we get

\[
(\omega - H_{nn})G_{nm} - H_{nn-1}G_{n-1m} - H_{nn+1}G_{n+1m} = \delta_{mn}.
\]

The matrix elements of the Hamiltonian \( H_{nn} \) that appear in this formula are 2×2 supermatrices (each principal layer contain two atomic layers) each of whose elements is a 10×10 matrix (since we are using a five wave functions basis by spin per atom). For example, for the surface

\[
H_{00} = \begin{pmatrix}
    h_{00} & h_{0-1} \\
    h_{-10} & h_{-1-1}
\end{pmatrix},
\]

\[
H_{01} = \begin{pmatrix}
    h_{0-2} & h_{0-3} \\
    h_{-1-2} & h_{-1-3}
\end{pmatrix}.
\]

Notice that the rows are labeled with the index of the surface principal layer zero (containing atomic layers 0 and 0), while the columns are indexed with the first principal and second principal layer (atomic layers 0 and -1 and atomic layers -2 and -3), respectively. We shall adopt the hypothesis of an ideal, nonreconstructed surface. Then, for the (001) surface, for example, we have one atomic layer of anions and one of cations per principal layer. Therefore, in this case \( h_{00} \neq h_{-1-1} \), \( h_{0-1} = h_{-10} \). Thus, to calculate \( H_{00} \) and \( H_{01} \), we need to know \( h_{00} \), \( h_{-1-1} \), \( h_{-10} \), and \( h_{-1-2} \). These matrices are readily written in a tight-binding language and can be calculated with the bulk parameters as mentioned above. They depend on the wave vector \( k \).

Using Eq. (4) for \( n = m \), and \( m = 0 \) for the surface, it is straightforward to get the surface Green's function\textsuperscript{12}

\[
G_{s}^{-1} = (\omega I - H_{00}) - H_{00}T
\]

and the principal layer projected bulk Green's function\textsuperscript{12}

\[
G_{b}^{-1} = G_{s}^{-1} - H_{10}T.
\]

It is customary to define the transfer matrices as

\[
G_{k+1p} = TG_{kp}, \quad k \geq p \geq 0,
\]

\[
G_{i+j+1} = T^{-1}G_{ij}, \quad j \geq i \geq 0.
\]

These matrices can be calculated by the quick algorithm of López-Sancho \textit{et al.},\textsuperscript{18} recalculated later by Baquero.\textsuperscript{19} (See Refs. 10 and 11 for a compilation of all the formulas.)

From the knowledge of the Green's function, the surface states, the surface resonances, and the surface-induced bulk states can be calculated from the poles of the real part of the corresponding Green's function. We have applied previously this formalism to other surfaces.\textsuperscript{5,16,25,27} We will now present our results.

III. RESULTS

The general characteristics of the zinc-blende II-VI semiconductor valence band as we have obtained from our calculations is as follows. The heavy-hole (hh) and light-hole (lh) bands follow each other closely in energy. The corresponding wave functions are mainly \((p_x, p_y)\) in character. The hh band disperses from \( \Gamma \) to \( X \) about 2.0 eV and the lh one about 2.4 eV. The spin-orbit splitting is around 1 eV in the Te compounds and around 0.5 in the Se ones. This band reaches \( X \) at about 5.0 eV. This is the band with the most dispersion and is composed essentially of \( p_z \) states only. Finally, a very deep bulk band of mostly \( s \) character appears below -10 eV. It disperses about 2 eV from \( \Gamma-X \). We will call this band \( b_{lo} \).

From the (001)-bulk-projected Green's function we also get the energy of the (001)-surface-induced bulk states. Three such states appear, \( B_{k1}, B_{l1}, \) and \( B_{s} \). These surface-induced bulk bands show no dispersion.\textsuperscript{5,26,29} \( B_{k1} \) was first found experimentally by Niles and Höchst\textsuperscript{3} and confirmed later by Gawlik \textit{et al.}\textsuperscript{4} for CdTe(001). \( B_{k1} \) mixes with the hh band at \( X \) and is located at the same energy (in \( \Gamma \)) as the surface resonance \((S_{k1}, \text{see below})\) of the cation-terminated (001) surface. \( B_{l1} \) mixes with
TABLE I. Characteristics of the band structure. The first two columns are the band-gap value (Γ6) and the spin-orbit splitting (Γ7), in the Γ point. The last three columns are the values at the X point of the Brillouin zone of the energy of the heavy-hole band (Xv), the light-hole band (Xv), and the spin orbit (Xv). All the values are in eV.

<table>
<thead>
<tr>
<th></th>
<th>Γ6</th>
<th>Γ7</th>
<th>Xv</th>
<th>Xv</th>
<th>Xv</th>
</tr>
</thead>
<tbody>
<tr>
<td>CdTe</td>
<td>1.602</td>
<td>-0.9</td>
<td>-1.7</td>
<td>-2.2</td>
<td>-4.4</td>
</tr>
<tr>
<td>CdSe</td>
<td>1.475</td>
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<td>-2.18</td>
<td>-2.36</td>
<td>-4.89</td>
</tr>
<tr>
<td>ZnTe</td>
<td>2.39</td>
<td>-0.91</td>
<td>-1.93</td>
<td>-2.40</td>
<td>-5.50</td>
</tr>
<tr>
<td>ZnSe</td>
<td>2.82</td>
<td>-0.45</td>
<td>-1.95</td>
<td>-2.19</td>
<td>-5.30</td>
</tr>
</tbody>
</table>

The lh band at X. The composition of both Bh and Bl is mainly of (pz, p0), while the one of Bx, which mixes with the spin-orbit band at X, is (s, pz). The three states appear at the same position in energy irrespective of the cation or anion termination of the surface as one expects for surface-induced bulk states that only depend on the surface through the boundary condition (the wave function has to be zero at the surface).

The (001)-surface valence band is rich in other features. In particular, three characteristic surface resonances do exist in this range of energy. Two correspond to the anion (Sx1, Sx2) and the other to the cation (Sx) termination of the (001) surface. In all the systems considered, the anion-terminated surface higher band (Sx1) follows roughly the dispersion of the hh bulk band but it is at slightly higher energy. The cation-terminated surface band (Sx) starts roughly around 2–3 eV from the top of the valence band in Γ and has a varying amount of dispersion. The two states appear at very different energy values and are distinctive of the termination of the surface for the four systems under consideration. One could speculate on the use of these two states to characterize the termination of the surface. A second (001)-surface anion resonance (Sx2) appears at much lower energies, for all the compounds considered, the nearest bulk band bx10. This surface state has no dispersion and its wave function is mostly of s character.

There is only one set of experimental results available at this moment to compare our results with, namely the already cited CdTe(001). We have studied this state and found that this is a surface-induced bulk one. Such bulk states appear as a consequence of the breaking of the symmetry implied by the creation of the (001) surface in the infinite medium. To support our argument, we showed specifically that a pole in the real part of the (001)-bulk-projected Green’s function appears at -4.4 eV from Γ to X, and does not appear either in the real part of the (001)-surface-projected Green’s function or in the real part of the bulk Green’s function for any termination (anion or cation) of the sample. In other words, this is neither a solution of the Schrödinger’s equation for an infinite medium nor a solution for a semi-infinite one that is localized at the surface. It is a bulk state existing as a consequence of the creation of the surface for which the crystal momentum is not a good quantum number anymore due to the new boundary condition. It has no ε(k) dispersion as a consequence. This is a surface-induced bulk state, as we already stated above.

In the rest of this paper, we want to describe in detail these (001) surface resonances and (001)-surface-induced bulk states for the four systems under consideration as well as the bulk bands obtained from our calculation. We will use our Hamiltonians to describe interfaces, superlattices, and quantum wells of these compounds in future work.

We summarize our results in Tables I–IV. In Table I, we give the characteristics of the bulk bands according to our obtained values. We give the gap and the spin-orbit coupling for each element and the value for the bands in Γ and at X. In Table II, we give the energy of the three surface-induced bulk states; in Table III, we give the energy of the surface resonances; finally, in Table IV, we give the composition of all the bands involved in our work.

A. CdTe(001)

Figure 1 shows the full valence band for this system. The bulk states appear as the dotted curves in the figure. Our tight-binding description reproduces correctly the band-gap value (1.6 eV) and the spin-orbit splitting (0.9 eV). Our tight-binding parameters were adjusted to give energy values at the X high-symmetry point of the Brillouin zone, which reproduce the experimental values

TABLE III. The energy position of the (001) surface resonances. All the values are in eV.

<table>
<thead>
<tr>
<th></th>
<th>Sx1</th>
<th>Sx2</th>
<th>Sx3</th>
</tr>
</thead>
<tbody>
<tr>
<td>CdTe</td>
<td>1.0</td>
<td>-1.2</td>
<td>-2.0</td>
</tr>
<tr>
<td>CdSe</td>
<td>1.0</td>
<td>-1.2</td>
<td>-2.0</td>
</tr>
<tr>
<td>ZnTe</td>
<td>1.0</td>
<td>-1.2</td>
<td>-2.0</td>
</tr>
<tr>
<td>ZnSe</td>
<td>1.0</td>
<td>-1.2</td>
<td>-2.0</td>
</tr>
</tbody>
</table>

TABLE IV. The wave function decomposition for all the states that appear in this study.

<table>
<thead>
<tr>
<th>State</th>
<th>Composition</th>
</tr>
</thead>
<tbody>
<tr>
<td>hh</td>
<td>(p0, p0)</td>
</tr>
<tr>
<td>lh</td>
<td>(p0, p0)</td>
</tr>
<tr>
<td>so</td>
<td>(p0)</td>
</tr>
<tr>
<td>Sx1</td>
<td>(s, p0)</td>
</tr>
<tr>
<td>Sx2</td>
<td>(s, p0)</td>
</tr>
<tr>
<td>Sx3</td>
<td>(s, p0)</td>
</tr>
</tbody>
</table>
FIG. 1. The full electronic valence-band structure for CdTe(001) in the Γ-X direction. The dot lines are the bulk band structure from direct diagonalization of the tight-binding Hamiltonian using the tight-binding parameters that appear in the Appendix. The two surface resonances for the anion-terminated (001) surface are denoted by full triangles, $S_{a1}$ and $S_{a2}$. The empty triangles are the surface resonance for the cation-terminated surface, $S_{c}$. The (001)-surface-induced bulk states $B_{h}$, $B_{l}$, and $B_{a}$ are the full dots (the dashed lines are a guide to the eye). Their occurrence is explained in the text. $B_{l}$ was first found experimentally by Niles and Höchst (Ref. 3).

for CdTe very closely.5,29 The heavy-hole and light-hole bands have a width of 1.8 eV and 2.2 for this interval of the Brillouin zone, respectively. The wave functions are constituted mainly by an admixture of $p_{x}$ and $p_{y}$ states. See Tables I and IV.

The surface resonances existing in this energy interval are identified as $S_{a1}$, $S_{a2}$, and $S_{c}$. The band $S_{a1}$ corresponds to the anion termination of the surface (black triangles); its width ($\Gamma-X$) is about 1 eV. The wave functions are constituted mainly by an admixture of $s$ and $p_{z}$ cation orbitals. The $S_{c}$ surface resonance (white triangles) corresponds to the cation-terminated (001) surface. This band develops from about 2.2 eV below the top of the valence band in $\Gamma$ to about $-3.5$ eV at $X$. Its width is therefore about 1.3 eV and is mainly of $s$ character. This band mixes with a surface-induced bulk band, $(B_{l})$, for about half of the wave-vector interval $\Gamma-X$ (see below). The $S_{a2}$ surface resonance shows no dispersion and is located at $-8.5$ eV. Its wave function is of $s$ character. These surface resonances were described before.27

Three surface-induced bulk states exist in this range of energy according to our findings. They are denoted by $B_{h}$, $B_{l}$, and $B_{a}$.

The states $B_{h}$ and $B_{l}$ are newly found surface-induced bulk states. $B_{h}$ is located at $-1.8$ eV and has no dispersion as is characteristic of these surface-induced bulk bands. $B_{h}$ seems to mix with the hh band at $X$. $B_{l}$ appears at $-2.2$ eV and mixes with the lh band at $X$. $B_{l}$ could present branches with slightly different energy and be actually a group of states differing slightly in energy. These states are mainly $(p_{x}, p_{y})$. $B_{l}$ starts at $\Gamma$ at the same energy as the surface resonance $S_{c}$. Their energy difference is very small for about half the interval $\Gamma-X$ when the surface resonance begins to have an important dispersion while the bulk one has none. We have obtained each state from the corresponding Green's function. The $B_{a}$ state has a $(s, p_{z})$ composition.

In conclusion, within the range of energy covered by its valence band, CdTe(001) presents, in addition to the known bulk bands, surface resonances and surface-induced bulk states. As is shown in Fig. 1, the anion terminated surface has two resonances $S_{a1}$ and $S_{a2}$ while the cation terminated one has only one surface resonance $S_{c}$. Furthermore, a total of three surface-induced bulk states appear, namely, $B_{h}$, $B_{l}$, and $B_{a}$. $B_{a}$ is the recently established surface-induced bulk state at $-4.4$ eV.3,25 There are all nondispersive states and appear at the same energy irrespective of the termination of the (001) surface.

B. CdSe(001)

In general, CdSe is grown in a hexagonal structure. Nevertheless, recent experiments31 have shown that it is possible to grow this material in a cubic phase and that it is stable. Our calculation has been done assuming the zinc-blende structure. Our bands do not apply for the hexagonal structure.

We can see in Fig. 2 the general characteristic of the II-VI zinc-blende semiconductor valence band already described above. The hh and lh bands follow each other closely in energy and are both composed by $(p_{x}, p_{y})$. The hh band disperses from $\Gamma$ to $X$ 2.2 eV and the lh one 2.4 eV. The spin-orbit splitting is 0.43 eV and the band $(p_{z})$ ends at $-5.0$ eV at $X$ and has therefore a width of about 4.6 eV. The $b_{10}$ band appears at $-11.0$ eV in $\Gamma$ and has little dispersion towards $X$.

For the anion-terminated (001) surface, there is one surface resonance $(S_{a1})$ of $(s, p_{x})$ composition that develops right from the top of the valence band in $\Gamma$ to $X$ with quite less dispersion than the hh band. At $X$ it takes the value 1.0 eV. The cation-terminated (001) surface shows a surface resonance $(S_{c})$ of $s$ character at a lower energy in $\Gamma$ $(<-2.2$ eV) that disperses very little up to half the $\Gamma-X$ interval but then disperses strongly and ends at $-4.0$ eV at $X$. Its width in the interval is 1.8 eV. The $S_{a2}$ band appears again very close to the $b_{10}$ bulk one and shows no dispersion since this is the general behavior for this surface resonance band.

The three (001)-surface-induced bulk states $(B_{h}$, $B_{l}$ and $B_{a}$) appear at $-2.2$ eV $(p_{x}, p_{y})$, $-2.45$ eV $(p_{x}, p_{y})$, and $-5.0$ eV $(s, p_{z})$ irrespective of the cation or anion termination of the surface. The first two mix with the hh and lh bands at $X$, respectively, and the third one mixes
with the spin-orbit band at $X$. None of them presents dispersion.

As we can conclude from the previous description, CdSe(001) follows very closely the general picture described above for the number, composition, and relative location of the surface resonances and surface-induced bulk states within the valence-band energy interval. We will now show that the last two compounds studied follow the same pattern.

**C. ZnTe(001)**

The full valence band for ZnTe(001) is presented in Fig. 3. It is clear that the general pattern is followed. $S_{s_1}$ is the highest band in energy. It starts at $\Gamma$ and disperses 1.0 eV as it reaches $X$. It is of $(s, p_x)$ character. The $hh$ and $lh$ bands [both $(p_x, p_y)$] follow each other and disperse 2.0 and 2.3 eV, respectively. $B_h$ and $B_l$ both mix with the $hh$ and $lh$ band, respectively, at $X$ and have the same composition. The $S_c$ resonance disperses 2.0 eV and mixes with the $B_h$ band at $\Gamma$. It is $s$-like character following the general rule. Finally, the $B_s$ state is located at $-5.3$ eV from the top of the valence band and is of $(s, p_x)$ character. This state seems to be composed of two states lying very closely in energy. The surface-induced bulk states present no disper-
D. ZnSe(001)

We present our results for this compound in Fig. 4. In all respects ZnSe(001) follows the general picture for the states in the valence-band energy interval. The hh, lh, and spin-orbit bands have the usual composition and behavior. Three different surface resonances appear, two for the anion surface (S_{01}, S_{02}) and one for the cation surface (S_{c}), and three surface-induced bulk states with the same usual characteristics. The surface resonances have been studied previously. We have summarized the details in Tables I–IV.

IV. CONCLUSIONS

In conclusion, we have studied the (001) surface resonances and surface-induced bulk states that appear in the range of the valence-band energy for the II-VI zinc-blende semiconductors. We found that a general pattern applies to the states and relative energy of these states with respect to the bulk bands. First, near the top of the valence band a surface resonance develops with little dispersion from Γ to X for the anion-terminated surface. A surface resonance also exists for the cation-terminated surface but it appears at a quite lower energy. A second anion surface resonance appears in the range of energies of the bottom of the valence band. Simultaneously three (001)-surface induced bulk states appear that show no dispersion and appear at the same energy irrespective of the surface termination. These states mix with the hh, lh, and spin-orbit bands at X, respectively.

A final comment is worth introducing here. We have considered ideal surfaces in this work. Anion-terminated zinc-blende (001) II-VI semiconductor surfaces can often reconstruct as is known to occur in the III-V systems. Rodriguez et al. found that in the anion-terminated surfaces [MTe(001); M=Zn, Cd, Hg], due to the possibility of occupancy of a donor state (of 5p-Te orbital composition) at 0 K, the reconstruction is to be expected. More recently Lu and Smith found by direct imaging of CdTe(001) surface using high-resolution electron microscopy that the (3×1) reconstruction, stabilized by Te atoms, involves the formation of surface dimers and the presence of vacancies. We have not considered these interesting phenomena in this work.

It would be interesting to study other directions of the surface as well as interfaces, superlattices, and quantum wells in different high-symmetry directions to characterize the different states that this break of symmetry due to the creation of a border might induce.

APPENDIX

We present in Table V the tight-binding parameters that we used for the systems studied in this work.

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**TABLE V. Tight-binding parameters used in our calculation. We used the notation of Bertho et al. (Ref. 25).**

<table>
<thead>
<tr>
<th>Parameter</th>
<th>CdTe(^{a})</th>
<th>ZnTe(^{b})</th>
<th>ZnSe(^{b})</th>
<th>CdSe(^{c})</th>
<th>Parameter</th>
<th>CdTe(^{a})</th>
<th>ZnTe(^{b})</th>
<th>ZnSe(^{b})</th>
<th>CdSe(^{c})</th>
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<td>-10.16740</td>
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<tr>
<td>$E_2$</td>
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<td>$V_{sp}$</td>
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<tr>
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<td>0.19373</td>
<td>0.14300</td>
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</tbody>
</table>

\(^{a}\)Reference 5.  
\(^{b}\)Reference 25.  
\(^{c}\)Reference 26.

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26. A. Camacho (private communication).