In region II, we represent the long-range (in R, at fixed r and γ) potential V_{LR} for the interaction between H_2 and H^- as [51]:

$$V_{LR}(R;r,\gamma) = D_{as}(r) + \frac{C_3^{\mathrm{th}}}{R^3} + \frac{C_4^{\mathrm{th}}}{R^4} \text{ with} \tag{1}$$
$$C_3^{\mathrm{th}} = -Q(r)P_2(\cos\gamma) \text{ and } C_4^{\mathrm{th}} = -\left[\alpha_0(r) + \alpha_2(r)P_2(\cos\gamma)\right]/2\,,$$

where the first term $D_{as}(r)$ is the sum of H^- and $H_2(r)$ energies at a given internuclear distance r of the H_2 molecule. The second term is the interaction between the electric charge of H^- with the quadrupole moment $Q(r)$ of H_2 (taken from Ref. [52]), and the third term is the interaction of the dipole moment of H_2 induced by H^-, involving the second order Legendre polynomial $P_2(\cos\gamma)$. The functions $\alpha_0(r)$ and $\alpha_2(r)$ are the isotropic and anisotropic polarizabilities of H_2, for which we used the analytical functions given in Ref. [11] that were obtained by fitting the numerical values from Ref. [53]. The dispersion energy varying as $1/R^6$ and other smaller terms are neglected, which is a good approximation because the long-range expansion is only used for $R > 56.4$ a.u.

In region I we used the following extrapolation formula in r for fixed R and γ:

$$V_{SR}(r;R,\gamma) = a(R,\gamma)e^{-b(R,\gamma)r}\,, \tag{2}$$

where $a(R,\gamma)$ and $b(R,\gamma)$ are functions of R and γ that are obtained considering the two *ab initio* energies $V(r=0.8;R,\gamma)$ and $V(r=1;R,\gamma)$ calculated at first two values of the coordinate r. In this way, we obtain the quantities a and b given on a two-dimensional grid of points in the (R,γ) space. Then we used the 2D B-spline interpolation to obtain smooth two-dimensional functions $a(R,\gamma)$ and $b(R,\gamma)$.

In region III, we extrapolate the PES in r and at fixed R and γ using a dispersion-like expression:

$$V_{LR}(r;R,\gamma) = D_0(R,\gamma) - \frac{C_6(R,\gamma)}{r^6}\,, \tag{3}$$

where the $D_0(R,\gamma)$ and $C_6(R,\gamma)$ (always positive) coefficients are obtained in a way similar to the coefficients a and b, considering the two last points $V(r=2.2\text{ a.u.};R,\gamma)$ and $V(r=2.4\text{ a.u.};R,\gamma)$. They are also interpolated using the 2D B-spline method for arbitrary values of R and γ.

The behavior of the PES in region IV is described by the short-range (in R) repulsive expression at given values of r and γ:

$$V_{SR}(R;r,\gamma) = A(r,\gamma)e^{-B(r,\gamma)R}\,, \tag{4}$$