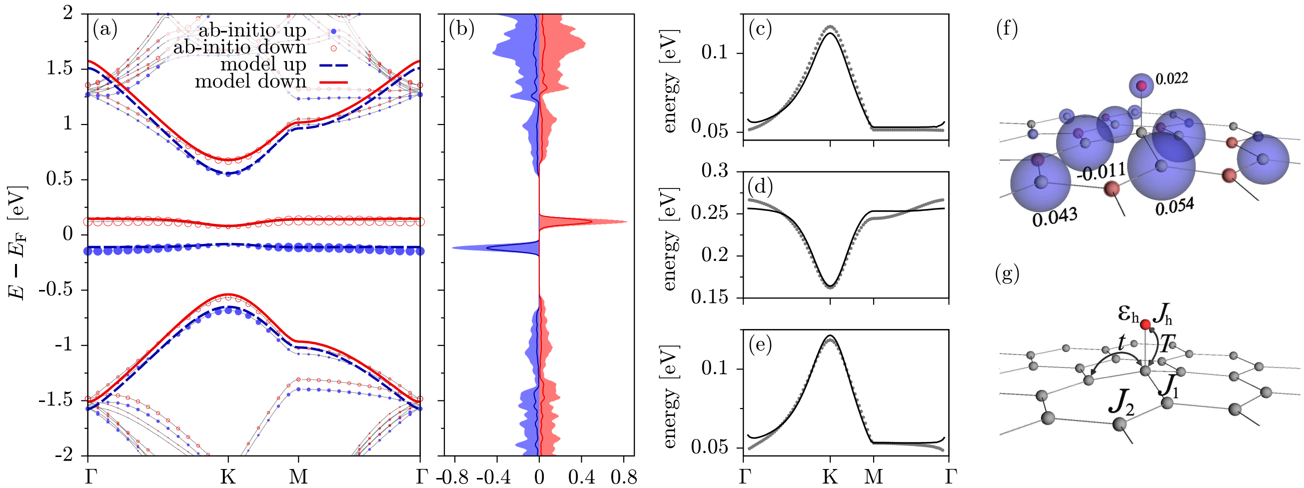

FIG. 1. First-principles results for a 5×5 graphene supercell with a single hydrogen adatom. (a) Spin-polarized band structure. The circles radii indicate the presence of p_z orbitals from the nearest neighbors to $\mathrm{C_H}$. Bold lines (dashed and solid) come from the exchange hopping model, Eq. (1). (b) Total density of states per atom (filled) and p_z projected local densities summed up to the third nearest carbon atoms to $\mathrm{C_H}$, normalized to the corresponding number of atoms in the set. Exchange splittings of the conduction (c), mid-gap (d), and valence (e) bands. Solid lines are from the model. (f) Local magnetic moments around hydrogen, indicated in μ_B. (g) Exchange hopping model of Eq. (1).

Spin relaxation rate.

The spin relaxation rate is obtained by formulating rate equations. Suppose we have an electron spin accumulation in graphene described by the spin (σ)-dependent chemical potential μ_σ. The electron distribution functions differ from equilibrium as $f_{k\sigma} = f_k^0 + \delta f_{k\sigma}$, where

$$\delta f_{k\sigma} \approx \left(-\frac{\partial f_k^0}{\partial \varepsilon_k}\right)(\mu_\sigma - \varepsilon_F). \tag{2}$$

We denoted as $f_k^0 = f^0(\varepsilon_k)$ the Fermi-Dirac function, ε_k the electron energy, and ε_F the Fermi level. The electron spin is

$$s = \sum_k \left(-\frac{\partial f_k^0}{\partial \varepsilon_k}\right)\mu_s, \tag{3}$$

with $\mu_s = \mu_\uparrow - \mu_\downarrow$. The spin relaxation rate is defined from equation,

$$\frac{\partial s}{\partial t} = \sum_k \left(-\frac{\partial f_k^0}{\partial \varepsilon_k}\right)\frac{\partial \mu_s}{\partial t} \equiv -\frac{s}{\tau_s}. \tag{4}$$

Let $W_{k\uparrow\Downarrow|k'\downarrow\Uparrow}$ be the spin-flip rate, that is, the rate of the transition of an electron with momentum k and spin up ($\uparrow$), in the presence of an impurity with spin down ($\Downarrow$), to another state of momentum k' and spin down ($\downarrow$), and impurity spin up ($\Uparrow$) (the electron and impurity spins are flipped). Let the probability of the impurity spin being $\Sigma = \{\Uparrow, \Downarrow\}$ be p_Σ. The rate equation for spin up electrons is

$$\begin{aligned}\frac{\partial f_{k\uparrow}}{\partial t} = &-\sum_{k'} W_{k\uparrow\Downarrow|k'\downarrow\Uparrow}\, p_\Downarrow f_{k\uparrow}(1 - f_{k'\downarrow}) \\ &+ \sum_{k'} W_{k'\downarrow\Uparrow|k\uparrow\Downarrow}\, p_\Uparrow f_{k'\downarrow}(1 - f_{k\uparrow}).\end{aligned} \tag{5}$$

Assuming unpolarized magnetic moments, $p_\Uparrow = p_\Downarrow = 1/2$, and using the symmetry of the spin-flip rates, we get

$$\frac{\partial f_{k\uparrow}}{\partial t} = -\frac{1}{2}\sum_{k'} W_{k\uparrow\Downarrow|k'\downarrow\Uparrow}(f_{k\uparrow} - f_{k'\downarrow}). \tag{6}$$

Similarly for the rate of $f_{k\downarrow}$. We can then write

$$\frac{\partial s}{\partial t} = -\sum_k \sum_{k'} W_{k\uparrow\Downarrow|k'\downarrow\Uparrow}(\delta f_{k\uparrow} - \delta f_{k'\downarrow}). \tag{7}$$

Substituting the spin accumulation and comparing with the defining equation for τ_s we get

$$\frac{1}{\tau_s} = \frac{\sum_k \sum_{k'} \left(-\partial f_k^0/\partial \varepsilon_k\right) W_{k\uparrow\Downarrow|k'\downarrow\Uparrow}}{\sum_k \left(-\partial f_k^0/\partial \varepsilon_k\right)}. \tag{8}$$

This equation is used to evaluate the temperature-dependent spin relaxation rates in the paper.

Finally, the transition rates in the presence of N_H impurities are calculated from the T-matrix,

$$W_{k\sigma\Sigma|k'\sigma'\Sigma'} = N_\mathrm{H}\frac{2\pi}{\hbar}\left|\mathrm{T}_{k\sigma\Sigma|k'\sigma'\Sigma'}\right|^2 \delta(\varepsilon_k - \varepsilon_{k'}). \tag{9}$$

All what is necessary for spin-flip rates is to transform the singlet and triplet T-matrix amplitudes, Eq. (4) in the paper, via composite spin states $|\uparrow, \Downarrow\rangle$ and $|\downarrow, \Uparrow\rangle$. This is a place where the function $f_{\sigma,\sigma'}(x, y)$, Eq. (7) in the main text, enters the game.