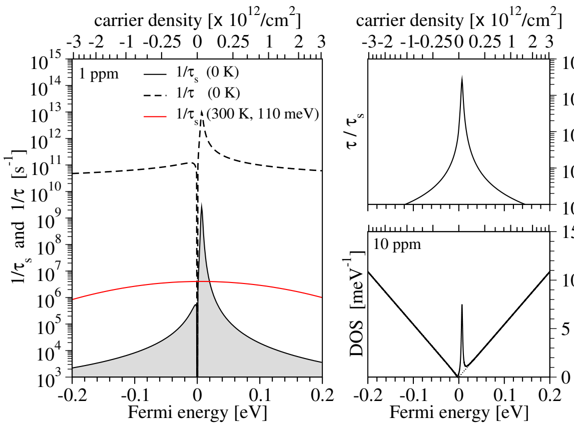


FIG. 5. Same as in Fig. 3 but for $J = -0.0004$ eV.

**Double-barrier atomic chain with a local moment sitting off the resonant site.**

In the paper we state that it is intuitively clear that the exchange coupling on the resonant site dominates spin-flip processes, so we can neglect $J$ on the carbon atoms near hydrogen. We can see this explicitly on our atomic chain model. Suppose a local spin giving exchange $J$ sits not inside resonant well but at one of the barriers, say the left one, i. e. $m = -1$. The model Hamiltonian is as follows:

$$H_{\rm off} = -t\sum_{\langle m,n\rangle}(c_m^\dagger c_n + c_n^\dagger c_m) + \sum_{m=\mp 1}\left(U c_m^\dagger c_m - J\delta_{m,-1}\,\hat{\mathbf{s}}\cdot\hat{\mathbf{S}}\right). \tag{10}$$

Transmissions and reflections amplitudes $\gamma_\ell$ and $\beta_\ell$ can be calculated to be,

$$\gamma_\ell = \frac{-e^{-ikb} + e^{ikb}\,\mathbb{Y}_\ell}{e^{ikb} + \frac{E_k}{t}(1 + e^{ikb}U/t) + e^{ikb}\,\mathbb{X}_\ell}\,, \tag{11}$$

$$\beta_\ell = \gamma_\ell\,\mathbb{X}_\ell - \mathbb{Y}_\ell\,, \tag{12}$$

where auxiliary functions $\mathbb{X}_\ell$ and $\mathbb{Y}_\ell$ are,

$$\mathbb{X}_\ell = \frac{t + e^{ikb}\,U}{t + e^{ikb}[U - (4\ell - 3)J]}\,, \tag{13}$$

$$\mathbb{Y}_\ell = \frac{t + e^{-ikb}\,[U - (4\ell - 3)J]}{t + e^{+ikb}[U - (4\ell - 3)J]}\,. \tag{14}$$

Figure 6 shows main characteristics of $H_{\rm off}$, Eq. (10), which allows us to call this model as off-resonant. Figure 6(a) provides the ratio $\mathcal{R}(E)$ for spin-flip *versus* spin-conserving probabilities, i. e.,

$$\mathcal{R}(E) = \left[\mathrm{t}(E)_{\sigma,\overline{\sigma}} + \mathrm{r}(E)_{\sigma,\overline{\sigma}}\right]/\left[\mathrm{t}(E)_{\sigma,\sigma} + \mathrm{r}(E)_{\sigma,\sigma}\right] \tag{15}$$

for different $J/t$. It is obvious when comparing with Fig. 2(a) in the main text that spin-flip processes are significantly suppressed. Figure 6(b) shows transmission and reflection probabilities $\mathrm{t}(E)_{\downarrow,\uparrow}$ and $\mathrm{r}(E)_{\downarrow,\uparrow}$ for

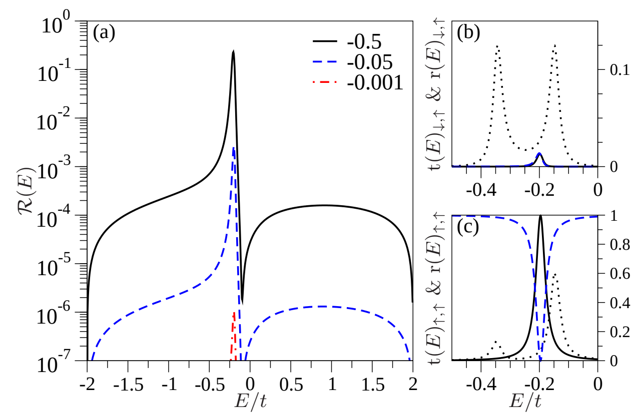


FIG. 6. Atomic chain with a local spin sitting at one of the barriers. (a) Ratio $\mathcal{R}(E)$ of spin-flip versus spin-conserving transition probabilities for $U/t = 10$ and indicated values of $J/t$. (b) Spin-flip transmission (solid) $\mathrm{t}(E)_{\downarrow,\uparrow}$ and reflection (dashed) $\mathrm{r}(E)_{\downarrow,\uparrow}$ for $J/t = -0.05$, both magnified by factor 10. (c) Same as in panel (b) but for spin-conserving quantities $\mathrm{t}(E)_{\uparrow,\uparrow}$ and $\mathrm{r}(E)_{\uparrow,\uparrow}$. For both panels (b) and (c) the dotted lines on the background show corresponding quantities from the resonant model, Eqs. (11) and (12) in the main paper.

$J/t = -0.05$ (to be visible, they are magnified by factor 10); the dotted line on the background is the transmission probability for the local moment on the resonant site inside the well (see Fig. 2(b) in the main text). Clearly, the spin-flip transitions are much more inhibited than in the resonant case discussed in the paper, justifying neglecting the off-resonant site exchange, especially when energy fluctuations from puddles wash out the fine structure of the resonant peaks. Figure 6(c) shows $\mathrm{t}(E)_{\uparrow,\uparrow}$ (solid line) and $\mathrm{r}(E)_{\uparrow,\uparrow}$ (dashed line) for $J/t = -0.05$. Similarly, the dotted line represents transmission probability $\mathrm{t}(E)_{\uparrow,\uparrow}$ in the resonant case.


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