Supplemental Material for Electrically driven singlet-triplet transition in graphene triangulene spin-1 chains

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A Derivatives BLBQ Hamiltonian

We start from the classical BLBQ Hamiltonian:

$$H = \frac{1}{2}\sum_{i\neq j}^{N} J_{ij}\left[\mathbf{S}_i\cdot\mathbf{S}_j + \beta_{ij}\left(\mathbf{S}_i\cdot\mathbf{S}_j\right)^2\right]. \tag{A.1}$$

The first derivate of Eq. A.1 respect to $\mathbf{S}_m$ is given by

$$\begin{aligned}\frac{\partial H}{\partial \mathbf{S}_m} &= \frac{1}{2}\sum_{i\neq j}^{N} J_{ij}\left(\delta_{im}\mathbf{S}_j + \delta_{jm}\mathbf{S}_i + 2\beta_{ij}\left(\mathbf{S}_i\cdot\mathbf{S}_j\right)\left[\delta_{im}\mathbf{S}_j + \delta_{jm}\mathbf{S}_i\right]\right) = \\ &= \sum_{j(j\neq m)} J_{jm}\left[\mathbf{S}_j + 2\beta_{jm}\left(\mathbf{S}_m\cdot\mathbf{S}_j\right)\mathbf{S}_j\right],\end{aligned} \tag{A.2}$$

and the second derivative is given by

$$\frac{\partial^2 H}{\partial \mathbf{S}_m\partial \mathbf{S}_n} = J_{mn}\left(1 + 2\beta_{mn}\left[\left(\mathbf{S}_m\cdot\mathbf{S}_n\right) + \mathbf{S}_n\circ\mathbf{S}_m\right]\right), \tag{A.3}$$

where $\circ$ is the dyadic product. Notice that the interaction energy between $\mathbf{S}_n$ and $\mathbf{S}_m$ is given by

$$\begin{aligned}\delta E^{(2)}_{nm} &= \delta\mathbf{S}_m\frac{\partial^2 H}{\partial \mathbf{S}_m\partial \mathbf{S}_n}\delta\mathbf{S}_n = \\ &= J_{nm}\left(\left(\delta\mathbf{S}_m\cdot\delta\mathbf{S}_n\right) + 2\beta_{nm}\left[\left(\delta\mathbf{S}_m\cdot\delta\mathbf{S}_n\right)\left(\mathbf{S}_m\cdot\mathbf{S}_n\right) + \left(\delta\mathbf{S}_m\cdot\mathbf{S}_n\right)\left(\mathbf{S}_m\cdot\delta\mathbf{S}_n\right)\right]\right),\end{aligned} \tag{A.4}$$

where $\delta\mathbf{S}_m$ is the infinitesimal variation of $\mathbf{S}_m$. In a collinear model ($\mathbf{S}_n \parallel \mathbf{S}_m$) we get that for transversal spin fluctuations $\delta\mathbf{S}_n\cdot\mathbf{S}_m = \mathbf{S}_n\cdot\delta\mathbf{S}_m = 0$ it is obtained

$$\delta E^{(2)}_{nm} = J_{nm}\left[1 + 2\beta_{nm}\left(\mathbf{S}_n\cdot\mathbf{S}_m\right)\right]\delta\mathbf{S}_n\cdot\delta\mathbf{S}_m. \tag{A.5}$$

B Singlet-Triplet Splitting

The energy splitting $\Delta E_{ST} = E(S=1) - E(S=0)$ of the singlet and triplet states lying deep in the Haldane gap can be obtained by quantizing the spin Hamiltonian (A.1). As we argue in the main text introducing a coupling J_{1N} between the spins on the edge of the chain a transition between the singlet and triplet state can be evoked. In Fig. 1 we show the energy splitting ΔE_{ST} in terms of J_{1N} for various chain lengths. It can be observed that the splitting is linear in the coupling. Also it is evident that longer chains require weaker coupling in order to drive them through the singlet-triplet transition. The energy scale of the singlet/triplet splitting can be tuned in the order of a couple of meVs, i. e., the splitting is easily resolved in experiments.

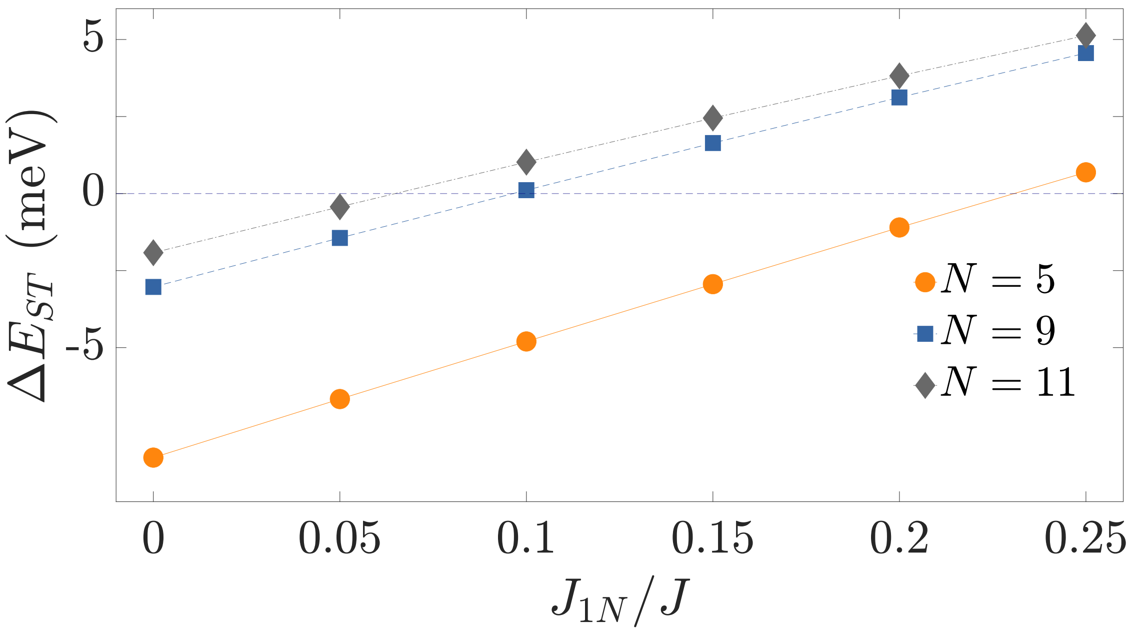

Figure 1: Singlet triplet energy-splitting as a function J_{1N}/J for $N = 5$, 9 and 11 chain lengths, all having $J = 19.75$ meV and $\beta = 0.05$.

C Electric Dipole on the Juntion

In the main text we state that that the electric tunability of the coupling constant J_{1N} is proportional to the dipole moment assotiated with the junction. In Fig. 2 we corroborate this statement by calculating the coupling constant in terms of the applied electric field and the calculated electric dipole. As it can be observed there is, to a good approximation, a linear relation between J_{1N} and the induced dipole moment $\mathcal{P}_y$. Performing a linear fit to the data points as function of $\mathcal{P}_y - \mathcal{P}_y^{(0)}$ where $\mathcal{P}_y^{(0)}$ is the electric dipole at zero external field, we obtain $J_{1N}/J = -0.038\ (e{\cdot}\text{Å})^{-1}\left(\mathcal{P}_y - \mathcal{P}_y^{(0)}\right) + 0.081$.

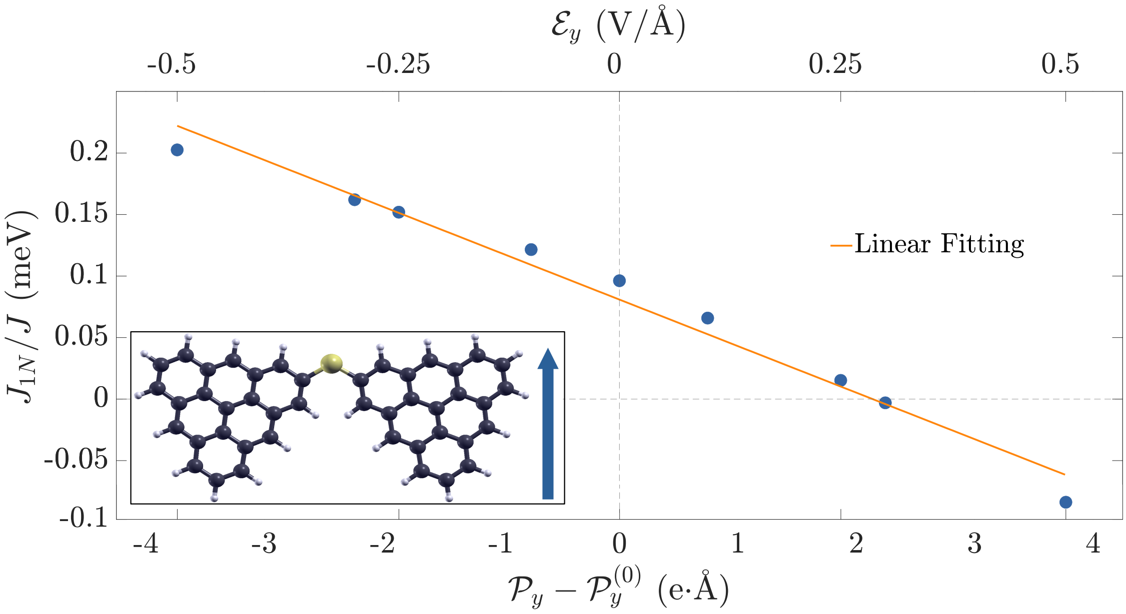

Figure 2: J_{1N} parameter as a function of the applied electric field and the total electric dipole on the junction.