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Supplementary information

Light-induced ferromagnetism in moiré superlattices

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Supplemental Information for

Light-Induced Ferromagnetism in Moiré Superlattices

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Supplementary Text: Calculation of exciton mediated magnetic exchange interaction.

Coulomb exchange between exciton and trapped hole. The Coulomb interactions between electrons, between holes, and between electron and hole are expressed as[1]

$$\begin{aligned} H_{ee} &= \frac{1}{2}\sum_{i,j,k,l}\langle ij|V|kl\rangle e_i^\dagger e_j^\dagger e_l e_k \\ H_{hh} &= \frac{1}{2}\sum_{\alpha,\beta,\gamma,\delta}\langle \alpha\beta|V|\gamma\delta\rangle h_\delta^\dagger h_\gamma^\dagger h_\alpha h_\beta \\ H_{eh} &= \sum_{i,j,\alpha,\beta}\left[\langle i\beta|V|\alpha j\rangle - \langle i\beta|V|j\alpha\rangle\right] e_i^\dagger h_\alpha^\dagger h_\beta e_j \end{aligned} \tag{1}$$

Here, $e_i^\dagger$ and $h_\alpha^\dagger = e_\alpha$ denote electron and hole creation operators, respectively. Their matrix elements between three-particle states consisting of one electron and two holes are

$$\begin{aligned} &\langle c_{\mathbf{e}'} v_{\mathbf{h}_1'} v_{\mathbf{h}_2'}|H_{hh}|c_{\mathbf{e}} v_{\mathbf{h}_1} v_{\mathbf{h}_2}\rangle \\ =\;& \frac{1}{2}\sum_{\alpha,\beta,\gamma,\delta}\langle \alpha\beta|V|\gamma\delta\rangle\langle 0\left|h_{\mathbf{h}_2'} h_{\mathbf{h}_1'} e_{\mathbf{e}'} h_\delta^\dagger h_\gamma^\dagger h_\alpha h_\beta e_{\mathbf{e}}^\dagger h_{\mathbf{h}_1}^\dagger h_{\mathbf{h}_2}^\dagger\right|0\rangle \\ =\;& \frac{1}{2}\sum_{\alpha,\beta,\gamma,\delta}\langle \alpha\beta|V|\gamma\delta\rangle\delta_{\mathbf{e}'\mathbf{e}}\langle 0\left|h_{\mathbf{h}_2'}\left(\delta_{\mathbf{h}_1'\delta} - h_\delta^\dagger h_{\mathbf{h}_1'}\right)h_\gamma^\dagger h_\alpha\left(\delta_{\mathbf{h}_1\beta} - h_{\mathbf{h}_1}^\dagger h_\beta\right)h_{\mathbf{h}_2}^\dagger\right|0\rangle \\ =\;& \frac{1}{2}\delta_{\mathbf{e}'\mathbf{e}}\left[\langle v_{\mathbf{h}_1} v_{\mathbf{h}_2}|V|v_{\mathbf{h}_1'} v_{\mathbf{h}_2'}\rangle + \langle v_{\mathbf{h}_2} v_{\mathbf{h}_1}|V|v_{\mathbf{h}_2'} v_{\mathbf{h}_1'}\rangle - \langle v_{\mathbf{h}_1} v_{\mathbf{h}_2}|V|v_{\mathbf{h}_2'} v_{\mathbf{h}_1'}\rangle - \langle v_{\mathbf{h}_2} v_{\mathbf{h}_1}|V|v_{\mathbf{h}_1'} v_{\mathbf{h}_2'}\rangle\right] \\ =\;& \delta_{\mathbf{e}'\mathbf{e}}\left(\langle v_{\mathbf{h}_1} v_{\mathbf{h}_2}|V|v_{\mathbf{h}_1'} v_{\mathbf{h}_2'}\rangle - \langle v_{\mathbf{h}_1} v_{\mathbf{h}_2}|V|v_{\mathbf{h}_2'} v_{\mathbf{h}_1'}\rangle\right) \end{aligned} \tag{2}$$

$$\begin{aligned} &\langle c_{\mathbf{e}'} v_{\mathbf{h}_1'} v_{\mathbf{h}_2'}|H_{eh}|c_{\mathbf{e}} v_{\mathbf{h}_1} v_{\mathbf{h}_2}\rangle \\ =\;& \sum_{\alpha,\beta,i,j}\left[\langle i\beta|V|\alpha j\rangle - \langle i\beta|V|j\alpha\rangle\right]\langle 0\left|h_{\mathbf{h}_2'} h_{\mathbf{h}_1'} e_{\mathbf{e}'} e_i^\dagger h_\alpha^\dagger h_\beta e_j e_{\mathbf{e}}^\dagger h_{\mathbf{h}_1}^\dagger h_{\mathbf{h}_2}^\dagger\right|0\rangle \\ =\;& \sum_{\alpha,\beta,i,j}\left[\langle i\beta|V|\alpha j\rangle - \langle i\beta|V|j\alpha\rangle\right]\delta_{\mathbf{e}'i}\delta_{\mathbf{e}j}\langle 0\left|h_{\mathbf{h}_2'}\left(\delta_{\mathbf{h}_1'\alpha} - h_\alpha^\dagger h_{\mathbf{h}_1'}\right)\left(\delta_{\mathbf{h}_1\beta} - h_{\mathbf{h}_1}^\dagger h_\beta\right)h_{\mathbf{h}_2}^\dagger\right|0\rangle \\ =\;& \left[\langle c_{\mathbf{e}'} v_{\mathbf{h}_1}|V|v_{\mathbf{h}_1'} c_{\mathbf{e}}\rangle - \langle c_{\mathbf{e}'} v_{\mathbf{h}_1}|V|c_{\mathbf{e}} v_{\mathbf{h}_1'}\rangle\right]\delta_{\mathbf{h}_2'\mathbf{h}_2} + \left[\langle c_{\mathbf{e}'} v_{\mathbf{h}_2}|V|v_{\mathbf{h}_2'} c_{\mathbf{e}}\rangle - \langle c_{\mathbf{e}'} v_{\mathbf{h}_2}|V|c_{\mathbf{e}} v_{\mathbf{h}_2'}\rangle\right]\delta_{\mathbf{h}_1'\mathbf{h}_1} \\ -\;& \left[\langle c_{\mathbf{e}'} v_{\mathbf{h}_2}|V|v_{\mathbf{h}_1'} c_{\mathbf{e}}\rangle - \langle c_{\mathbf{e}'} v_{\mathbf{h}_2}|V|c_{\mathbf{e}} v_{\mathbf{h}_1'}\rangle\right]\delta_{\mathbf{h}_2'\mathbf{h}_1} - \left[\langle c_{\mathbf{e}'} v_{\mathbf{h}_1}|V|v_{\mathbf{h}_2'} c_{\mathbf{e}}\rangle - \langle c_{\mathbf{e}'} v_{\mathbf{h}_1}|V|c_{\mathbf{e}} v_{\mathbf{h}_2'}\rangle\right]\delta_{\mathbf{h}_1'\mathbf{h}_2} \end{aligned} \tag{3}$$

Here the bold character denotes all the degrees of freedom including momentum $\mathbf{k}$ and spin-valley index σ, e.g. $\mathbf{e} \equiv (\mathbf{k}_e, \sigma_e)$). $\langle c_{\mathbf{k}_1,\sigma_1} v_{\mathbf{k}_2,\sigma_2}|V|c_{\mathbf{k}_3,\sigma_3} v_{\mathbf{k}_4,\sigma_4}\rangle$ and $\langle v_{\mathbf{k}_1,\sigma_1} v_{\mathbf{k}_2,\sigma_2}|V|v_{\mathbf{k}_3,\sigma_3} v_{\mathbf{k}_4,\sigma_4}\rangle$ are the two-body matrix elements in momentum space with the following expression[2]

$$\begin{aligned} &\langle c_{\mathbf{k}_1,\sigma_1} v_{\mathbf{k}_2,\sigma_2}|V|c_{\mathbf{k}_3,\sigma_3} v_{\mathbf{k}_4,\sigma_4}\rangle = \int d\mathbf{r}_1 d\mathbf{r}_2 \psi^*_{\mathbf{k}_1,\sigma_1,c}(\mathbf{r}_1)\psi_{\mathbf{k}_2,\sigma_2,v}(\mathbf{r}_2)V(\mathbf{r}_1 - \mathbf{r}_2)\psi_{\mathbf{k}_3,\sigma_3,c}(\mathbf{r}_1)\psi^*_{\mathbf{k}_4,\sigma_4,v}(\mathbf{r}_2) \\ =\;& \delta_{\mathbf{k}_1-\mathbf{k}_3,\mathbf{k}_2-\mathbf{k}_4}\sum_{\mathbf{G}}\frac{V(\mathbf{k}_1-\mathbf{k}_3+\mathbf{G})}{A}\langle u_{\mathbf{k}_1,\sigma_1,c}|e^{i\mathbf{G}\cdot\mathbf{r}}|u_{\mathbf{k}_3,\sigma_3,c}\rangle\langle u_{\mathbf{k}_2,\sigma_2,v}|e^{-i\mathbf{G}\cdot\mathbf{r}}|u_{\mathbf{k}_4,\sigma_4,v}\rangle \\ &\langle v_{\mathbf{k}_1,\sigma_1} v_{\mathbf{k}_2,\sigma_2}|V|v_{\mathbf{k}_3,\sigma_3} v_{\mathbf{k}_4,\sigma_4}\rangle = \int d\mathbf{r}_1 d\mathbf{r}_2 \psi_{\mathbf{k}_1,\sigma_1,v}(\mathbf{r}_1)\psi_{\mathbf{k}_2,\sigma_2,v}(\mathbf{r}_2)V(\mathbf{r}_1 - \mathbf{r}_2)\psi^*_{\mathbf{k}_3,\sigma_3,v}(\mathbf{r}_1)\psi^*_{\mathbf{k}_4,\sigma_4,v}(\mathbf{r}_2) \\ =\;& \delta_{\mathbf{k}_1+\mathbf{k}_2,\mathbf{k}_3+\mathbf{k}_4}\sum_{\mathbf{G}}\frac{V(\mathbf{k}_1-\mathbf{k}_3+\mathbf{G})}{A}\langle u_{\mathbf{k}_3,\sigma_3,v}|e^{i\mathbf{G}\cdot\mathbf{r}}|u_{\mathbf{k}_1,\sigma_1,v}\rangle\langle u_{\mathbf{k}_4,\sigma_4,v}|e^{-i\mathbf{G}\cdot\mathbf{r}}|u_{\mathbf{k}_2,\sigma_2,v}\rangle \end{aligned} \tag{4}$$

where $|u\rangle$ is the periodical part of the Bloch function, $V(\mathbf{k}) = \int d\mathbf{r} e^{i\mathbf{k}\cdot\mathbf{r}}V(\mathbf{r})$ is the Fourier transformation of 2D Coulomb potential $V(\mathbf{r})$ and A is the area of the 2D plane (for box normalization).

The Coulomb exchange interaction between the exciton and the trapped hole is evaluated, in the basis $|\Psi_{\mathbf{k},j\sigma_e\sigma_h\sigma}\rangle = d_{j\sigma}^\dagger a_{\mathbf{k}\sigma_e\sigma_h}^\dagger|0\rangle$, where $|\Psi_{X,\mathbf{k}\sigma_e\sigma_h}\rangle = a_{\mathbf{k}\sigma_e\sigma_h}^\dagger|0\rangle$ is the momentum eigenstate of the interlayer exciton with electron (hole) spin/valley index σ_e (σ_h), and $|\Psi_{h,j\sigma}\rangle = d_{j\sigma}^\dagger|0\rangle$ denotes a trapped hole wave packet centered at $\mathbf{R}_j$. We have $|\Psi_{X,\mathbf{k}\sigma_e\sigma_h}\rangle = \sum_{\mathbf{q}_X}$

$\psi_X(\mathbf{q}_X) e^{\dagger}_{\mathbf{q}_X+\mathbf{k}/2,\sigma_e} h^{\dagger}_{\mathbf{q}_X-\mathbf{k}/2,\sigma_h}|0\rangle$ and $|\Psi_{h,j\sigma}\rangle = \sum_{\mathbf{k}_h} \mathrm{e}^{i\mathbf{k}_h\cdot\mathbf{R}_j} W_h(\mathbf{k}_h) h^{\dagger}_{\mathbf{k}_h,\sigma}|0\rangle$ with $W_h(\mathbf{r}_h) = e^{-r_h^2/a_h^2}$ and $\psi_X(\mathbf{r}_{eh}) = e^{-r_{eh}/a_b}$ for the 1s exciton. The corresponding Fourier transformations are $W_h(\mathbf{k}_h) = \frac{a_h^2}{2} e^{-\frac{1}{4}a_h^2 k_h^2}$ and $\psi_X(\mathbf{q}_X) = \frac{a_b^2}{(1+q_X^2 a_b^2)^{3/2}}$. The normalized wave function of the basis state is:

$$|\Psi_{\mathbf{k},j\sigma_e\sigma_h\sigma}\rangle = C\sum_{\mathbf{q}_X,\mathbf{k}_h} \psi_X(\mathbf{q}_X) \mathrm{e}^{i\mathbf{k}_h\cdot\mathbf{R}_j} W_h(\mathbf{k}_h) e^{\dagger}_{\mathbf{q}_X+\mathbf{k}/2,\sigma_e} h^{\dagger}_{\mathbf{q}_X-\mathbf{k}/2,\sigma_h} h^{\dagger}_{\mathbf{k}_h,\sigma}|0\rangle \tag{5}$$

with the normalization condition:

$$\langle\Psi_{\mathbf{k},j\sigma_e\sigma_h\sigma}|\Psi_{\mathbf{k},j\sigma_e\sigma_h\sigma}\rangle = \frac{C^2A^2}{(2\pi)^4}\int d\mathbf{q}_X d\mathbf{k}_h |W_h(\mathbf{k}_h)|^2 |\psi_X(\mathbf{q}_X)|^2 = 1 \tag{6}$$

For the interlayer exciton and trapped hole in the type-II heterobilayer, holes are layer separated from the electron component of the exciton. Therefore, we can neglect the terms that relies on wavefunction overlap between the electron and hole (second, fourth, sixth and eighth term in Eq. (3)). The Coulomb exchange interaction between the exciton and the trapped hole are then contributed by the second term in Eq. (2) from the hole-hole interaction, and the fifth and seventh terms in Eq. (3) from the electron-hole interaction,

$$\begin{aligned}
&\left\langle\Psi_{\mathbf{k}',j\sigma_e'\sigma_h'\sigma'}\right| H_{hh}\left|\Psi_{\mathbf{k},j\sigma_e\sigma_h\sigma}\right\rangle_{second} \\
= \; & -\delta_{\sigma_e'\sigma_e} C^2 \sum_{\mathbf{q}_X\mathbf{k}_h\mathbf{q}_X'\mathbf{k}_h'} \mathrm{e}^{i(\mathbf{k}_h-\mathbf{k}_h')\cdot\mathbf{R}_j} W_h^*(\mathbf{k}_h')\psi_X^*(\mathbf{q}_X') W_h(\mathbf{k}_h)\psi_X(\mathbf{q}_X)\delta_{\mathbf{q}_X'+\mathbf{k}'/2,\mathbf{q}_X+\mathbf{k}/2} \\
& \delta_{\mathbf{k}'-\mathbf{k}_h',\mathbf{k}-\mathbf{k}_h} \sum_{\mathbf{G}} \frac{V(\mathbf{q}_X-\mathbf{k}/2-\mathbf{k}_h'+\mathbf{G})}{A} \left\langle u_{\mathbf{k}_h',v}\right| \mathrm{e}^{i\mathbf{G}\cdot\mathbf{r}}\left|u_{\mathbf{q}_X-\mathbf{k}/2,v}\right\rangle \left\langle u_{\mathbf{q}_X'-\mathbf{k}'/2,v}\right| \mathrm{e}^{-i\mathbf{G}\cdot\mathbf{r}}\left|u_{\mathbf{k}_h,v}\right\rangle \\
\approx \; & -\delta_{\sigma_e'\sigma_e}\delta_{\sigma_h'\sigma}\delta_{\sigma_h\sigma'} \mathrm{e}^{-i(\mathbf{k}-\mathbf{k}')\cdot\mathbf{R}} C^2/A \\
& \sum_{\mathbf{q}_X\mathbf{k}_h} W_h^*(\mathbf{k}_h-\mathbf{k}+\mathbf{k}')\psi_X^*\left(\mathbf{q}_X+\frac{\mathbf{k}-\mathbf{k}'}{2}\right) W_h(\mathbf{k}_h)\psi_X(\mathbf{q}_X) V\left(\mathbf{q}_X+\frac{\mathbf{k}}{2}-\mathbf{k}'-\mathbf{k}_h\right)
\end{aligned} \tag{7}$$

$$\begin{aligned}
&\left\langle\Psi_{\mathbf{k}',j\sigma_e'\sigma_h'\sigma'}\right| H_{eh}\left|\Psi_{\mathbf{k},j\sigma_e\sigma_h\sigma}\right\rangle_{fifth} \approx \delta_{\sigma_e'\sigma_e}\delta_{\sigma_h'\sigma}\delta_{\sigma_h\sigma'} \mathrm{e}^{-i(\mathbf{k}-\mathbf{k}')\cdot\mathbf{R}} C^2/A \\
&\sum_{\mathbf{q}_X'\mathbf{k}_h'} W_h^*(\mathbf{k}_h')\psi_X^*(\mathbf{q}_X') W_h(\mathbf{k}_h'+\mathbf{k}-\mathbf{k}')\psi_X\left(\mathbf{k}_h'+\frac{\mathbf{k}}{2}\right) V\left(\mathbf{q}_X'+\frac{\mathbf{k}'}{2}-\mathbf{k}-\mathbf{k}_h'\right)
\end{aligned} \tag{8}$$

$$\begin{aligned}
&\left\langle\Psi_{\mathbf{k}',j\sigma_e'\sigma_h'\sigma'}\right| H_{eh}\left|\Psi_{\mathbf{k},j\sigma_e\sigma_h\sigma}\right\rangle_{seventh} \approx \delta_{\sigma_e'\sigma_e}\delta_{\sigma_h'\sigma}\delta_{\sigma_h\sigma'} \mathrm{e}^{-i(\mathbf{k}-\mathbf{k}')\cdot\mathbf{R}} C^2/A \\
&\sum_{\mathbf{q}_X\mathbf{k}_h} W_h^*(\mathbf{k}_h+\mathbf{k}'-\mathbf{k})\psi_X^*\left(\mathbf{k}_h+\frac{\mathbf{k}'}{2}\right) W_h(\mathbf{k}_h)\psi_X(\mathbf{q}_X) V\left(\mathbf{q}_X+\frac{\mathbf{k}}{2}-\mathbf{k}'-\mathbf{k}_h\right)
\end{aligned} \tag{9}$$

Exciton mediated spin-spin interaction between trapped holes. The Hamiltonian for the exciton and hole in the basis of exciton momentum eigenstate and moiré trapped hole is,

$$\begin{aligned}
H_0 &= \sum_{\mathbf{k}\sigma_e\sigma_h} \epsilon_{\mathbf{k}\sigma_e\sigma_h} a^{\dagger}_{\mathbf{k}\sigma_e\sigma_h} a_{\mathbf{k}\sigma_e\sigma_h} + \sum_{j\sigma} \epsilon_d d^{\dagger}_{j\sigma} d_{j\sigma} \\
H_v &= \sum_{j\mathbf{k}\mathbf{k}'\sigma\sigma'\sigma_e} \langle\Psi_{\mathbf{k},j\sigma_e\sigma\sigma'}| H_{Coulomb} |\Psi_{\mathbf{k}',j\sigma_e\sigma'\sigma}\rangle d^{\dagger}_{j\sigma'} d_{j\sigma} a^{\dagger}_{\mathbf{k}\sigma_e\sigma} a_{\mathbf{k}'\sigma_e\sigma'} \\
&= \sum_{j\mathbf{k}\mathbf{k}'\sigma\sigma'\sigma_e} I\, e^{-i(\mathbf{k}-\mathbf{k}')\cdot\mathbf{R}_j} d^{\dagger}_{j\sigma'} d_{j\sigma} a^{\dagger}_{\mathbf{k}\sigma_e\sigma} a_{\mathbf{k}'\sigma_e\sigma'}
\end{aligned} \tag{10}$$

where only the exchange part of the exciton hole interaction is included as H_v. The exciton mediated spin-spin interaction between trapped holes arises as the second order perturbation of the off-diagonal exciton-hole exchange. And the exciton-hole exchange coefficient I in H_v contains three contributions given in Eq. (7), (8) and (9) respectively. The term from the intralayer hole-hole interaction is given as,

$$
\begin{aligned}
I_{hh}A &\approx -\frac{64\pi^2}{A^2 a_b^2 a_h^2}\sum_{\mathbf{q}_X\mathbf{k}_h} W_h^*(\mathbf{k}_h-\mathbf{k}+\mathbf{k}')\psi_X^*\left(\mathbf{q}_X+\frac{\mathbf{k}-\mathbf{k}'}{2}\right)W_h(\mathbf{k}_h)\psi_X(\mathbf{q}_X)V\left(\mathbf{q}_X+\frac{\mathbf{k}}{2}-\mathbf{k}'-\mathbf{k}_h\right) \\
&\approx -\frac{4}{\pi^2 a_b^2 a_h^2}\int d\mathbf{q}_X d\mathbf{k}_h W_h^*(\mathbf{k}_h)\psi_X^*(\mathbf{q}_X)W_h(\mathbf{k}_h)\psi_X(\mathbf{q}_X)\frac{e^2}{4\pi\varepsilon_r\varepsilon_0(k_{TF}+|\mathbf{q}_X-\mathbf{k}_h|)}
\end{aligned}
\tag{11}
$$

In the second line, the dependence on $\mathbf{k}$ and $\mathbf{k}'$is neglected, because the exciton states involved are the small momentum ones (much smaller compared to the momentum space width of W_h and ψ_X). In such limit, both terms from the interlayer electron-hole interaction have the same form,

$$
\mathrm{I}_{\mathrm{eh}}A \approx \frac{4}{\pi^2 a_b^2 a_h^2}\int d\mathbf{q}_X d\mathbf{k}_h W_h^*(\mathbf{k}_h)\psi_X^*(\mathbf{k}_h)W_h(\mathbf{k}_h)\psi_X(\mathbf{q}_X)\frac{e^2}{4\pi\varepsilon_r\varepsilon_0(k_{TF}+|\mathbf{q}_X-\mathbf{k}_h|)}e^{-d|\mathbf{q}_X-\mathbf{k}_h|}
\tag{12}
$$

With the selection of the following parameters, $a_b = 2$ nm, $a_h = 2$ nm, $k_{TF} = \frac{10\omega_0}{c} \approx \frac{100}{7}\ \mu\mathrm{m}^{-1}$, $d = 0.7$ nm and $\varepsilon_r = 4$ (dielectric constant at exciton binding energy), we obtain $I_{hh}\mathrm{A} \approx -0.71\ \mathrm{eV}\cdot\mathrm{nm}^2$ and $I_{eh}\mathrm{A} \approx 0.5\ \mathrm{eV}\cdot\mathrm{nm}^2$. Thus,

$$
I = I_{hh} + 2I_{eh} \approx \frac{0.29\ \mathrm{eV}\cdot\mathrm{nm}^2}{A}
\tag{13}
$$

To find the form of the exciton mediated spin-spin interaction between trapped holes, Schrieffer-Wolff transformation is used to eliminate the off-diagonal H_v. Introducing,

$$
S = -\sum_{j\mathbf{k}\mathbf{k}'\sigma\sigma'\sigma_e}\frac{Ie^{-i(\mathbf{k}-\mathbf{k}')\cdot\mathbf{R}_j}}{\epsilon_{\mathbf{k}\sigma_e\sigma}-\epsilon_{\mathbf{k}'\sigma_e\sigma'}}d_{j\sigma'}^\dagger d_{j\sigma}a_{\mathbf{k}\sigma_e\sigma}^\dagger a_{\mathbf{k}'\sigma_e\sigma'}
\tag{14}
$$

which satisfies $[S, H_0] = -H_v$, the leading perturbation term in the transformed Hamiltonian $e^S He^{-S}$ is then given as $\Delta H^{(2)} = \frac{1}{2}[S, H_v]$,

$$
\Delta H^{(2)} = \sum_{jj'\mathbf{k}\mathbf{k}'\sigma\sigma'\sigma_e}\frac{I^2}{2}\frac{e^{i(\mathbf{k}'-\mathbf{k})\cdot(\mathbf{R}_j-\mathbf{R}_{j'})}}{\epsilon_{\mathbf{k}\sigma_e\sigma}-\epsilon_{\mathbf{k}'\sigma_e\sigma'}}\left[d_{j\sigma'}^\dagger d_{j\sigma}a_{\mathbf{k}\sigma_e\sigma}^\dagger a_{\mathbf{k}'\sigma_e\sigma'}, d_{j'\sigma}^\dagger d_{j'\sigma'}a_{\mathbf{k}'\sigma_e\sigma'}^\dagger a_{\mathbf{k}\sigma_e\sigma}\right]
\tag{15}
$$

Tracing out the exciton degrees of freedom with a diagonal density matrix, i.e. $\langle a_{\mathbf{k}\sigma_e\sigma}^\dagger a_{\mathbf{k}\sigma_e\sigma}\rangle = f_{\mathbf{k}\sigma_e\sigma}$,

$$
\begin{aligned}
\Delta H^{(2)} = &\sum_{j\mathbf{k}\mathbf{k}'\sigma\sigma'\sigma_e}\frac{I^2}{2}\frac{1}{\epsilon_{\mathbf{k}\sigma_e\sigma}-\epsilon_{\mathbf{k}'\sigma_e\sigma'}}\left(f_{\mathbf{k}\sigma_e\sigma}d_{j\sigma'}^\dagger d_{j\sigma'} - f_{\mathbf{k}'\sigma_e\sigma'}d_{j\sigma}^\dagger d_{j\sigma}\right) \\
&+\sum_{j\mathbf{k}\mathbf{k}'\sigma\sigma'\sigma_e}\frac{I^2}{2}\frac{1}{\epsilon_{\mathbf{k}\sigma_e\sigma}-\epsilon_{\mathbf{k}'\sigma_e\sigma'}}\left(f_{\mathbf{k}\sigma_e\sigma}f_{\mathbf{k}'\sigma_e\sigma'}d_{j\sigma'}^\dagger d_{j\sigma'} - f_{\mathbf{k}'\sigma_e\sigma'}f_{\mathbf{k}\sigma_e\sigma}d_{j\sigma}^\dagger d_{j\sigma}\right) \\
&+\sum_{jj'\mathbf{k}\mathbf{k}'\sigma\sigma'\sigma_e}\frac{I^2}{2}\frac{e^{i(\mathbf{k}'-\mathbf{k})\cdot(\mathbf{R}_j-\mathbf{R}_{j'})}}{\epsilon_{\mathbf{k}\sigma_e\sigma}-\epsilon_{\mathbf{k}'\sigma_e\sigma'}}\left(f_{\mathbf{k}\sigma_e\sigma}-f_{\mathbf{k}'\sigma_e\sigma'}\right)d_{j\sigma'}^\dagger d_{j\sigma}d_{j'\sigma}^\dagger d_{j'\sigma'}
\end{aligned}
\tag{16}
$$

where the last term is the mediated spin-spin interaction between the holes (as illustrated in Fig. 3d). We note that the exchange with the trapped hole can flip an exciton between the spin triplet and singlet configurations (of the intravalley and intervalley configurations respectively in the R type WS_2/WSe_2). So, the denominator involves the energy of the species $\epsilon_{\mathbf{k}\sigma_e\sigma} - \epsilon_{\mathbf{k}'\sigma_e\sigma'}$. As electron-hole exchange is quenched by layer separation, there is no splitting between the intravalley spin triplet and intervalley singlet exciton dispersion, i.e. $\epsilon_{\mathbf{k}} \equiv \epsilon_{\mathbf{k}\sigma\sigma} = \epsilon_{\mathbf{k}\sigma\bar{\sigma}}$. Thus, the spin-spin interaction terms in $\Delta H^{(2)}$ between the trapped holes is of the form,

$$H_{SS} = \sum_{jj'} J\left(\mathbf{R}_j - \mathbf{R}_{j'}\right)\mathbf{S}_j \cdot \mathbf{S}_{j'} \tag{17}$$

where

$$J\left(\mathbf{R}_j - \mathbf{R}_{j'}\right) \quad = \sum_{\mathbf{k}\mathbf{k}'} \frac{I^2}{2} \cos\left[(\mathbf{k}' - \mathbf{k}) \cdot \left(\mathbf{R}_j - \mathbf{R}_{j'}\right)\right] \frac{f_{\mathbf{k}} - f_{\mathbf{k}'}}{\epsilon_{\mathbf{k}} - \epsilon_{\mathbf{k}'}} \tag{18}$$

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