I. POTENTIAL MATRIX

From Kohn-Sham equations, we can write the Kohn-Sham potential matrix as the following equation.

$$\underline{\underline{v}}^{\mathrm{KS}}(\mathbf{r}) = \sum_{j=0}^{3} \sigma_j V_j^{\mathrm{KS}}(\mathbf{r}) \tag{1}$$

Defining V_j^{KS} as a vector made up of the potentials in the system, B_x, B_y, and B_z, we can find the $\underline{\underline{v}}_j^{\mathrm{KS}}$ by carrying out our multiplication of the Pauli matrices on V_j^{KS}.

$$\underline{\underline{v}}^{\mathrm{KS}}(\mathbf{r}) = \sigma_0 V_0^{\mathrm{KS}} + \sigma_1 V_1^{\mathrm{KS}} + \sigma_2 V_2^{\mathrm{KS}} + \sigma_3 V_3^{\mathrm{KS}}$$

$$\underline{\underline{v}}^{\mathrm{KS}}(\mathbf{r}) = \begin{pmatrix} 1 & 0 \\ 0 & 1 \end{pmatrix} V_0^{\mathrm{KS}} + \begin{pmatrix} 0 & 1 \\ 1 & 0 \end{pmatrix} V_1^{\mathrm{KS}} + \begin{pmatrix} 0 & -i \\ i & 0 \end{pmatrix} V_2^{\mathrm{KS}} + \begin{pmatrix} 1 & 0 \\ 0 & -1 \end{pmatrix} V_3^{\mathrm{KS}}$$

Then we combine them altogether, replacing our V_j potentials with the actual potentials.

$$\underline{\underline{v}}^{\mathrm{KS}}(\mathbf{r}) = \begin{pmatrix} V + B_z & B_x - iB_y \\ B_x + iB_y & V - B_z \end{pmatrix}$$

Now, we need to determine the derivatives of our wavefunctions with respect to V_j.

$$|\delta\phi_n\rangle = \sum_{k\neq j} \frac{\langle\phi_k|\,\delta V\,|\phi_n\rangle}{E_n^{(0)} - E_k^{(0)}}\,|\phi_k\rangle \tag{2}$$

Using our spinor notation, we can reorganize our equation to get our perturbation equation with equation.

$$\begin{pmatrix} \delta\phi_{i\uparrow} \\ \delta\phi_{i\downarrow} \end{pmatrix} = \sum_n \sum_{j\neq i} \frac{\begin{pmatrix} \phi_{j\uparrow}^* & \phi_{j\downarrow}^* \end{pmatrix} \underline{\underline{v}}^{\mathrm{KS}} \begin{pmatrix} \phi_{i\uparrow} \\ \phi_{i\downarrow} \end{pmatrix}}{E_i - E_j} \begin{pmatrix} \phi_{j\uparrow} \\ \phi_{j\downarrow} \end{pmatrix}$$

$$\begin{pmatrix} \delta\phi_{i\uparrow} \\ \delta\phi_{i\downarrow} \end{pmatrix} = \sum_n \sum_{j\neq i} \frac{\begin{pmatrix} \phi_{j\uparrow}^* & \phi_{j\downarrow}^* \end{pmatrix} \begin{pmatrix} V + B_z & B_x - iB_y \\ B_x + iB_y & V - B_z \end{pmatrix} \begin{pmatrix} \phi_{i\uparrow} \\ \phi_{i\downarrow} \end{pmatrix}}{E_i - E_j} \begin{pmatrix} \phi_{j\uparrow} \\ \phi_{j\downarrow} \end{pmatrix} \tag{3}$$

Multiplying this matrix equation out, we can explicitly define the variables we are deriving with respect to.

$$\begin{pmatrix} \delta\phi_{i\uparrow} \\ \delta\phi_{i\downarrow} \end{pmatrix} = \sum_{j\neq i} \left\{ \Big([V_0 + V_3]\phi_{i\uparrow} + [V_1 + iV_2]\phi_{i\downarrow} \Big)\phi_{j\uparrow} + \Big([V_1 + iV_2]\phi_{i\uparrow} + [V_0 - iV_3]\phi_{i\downarrow} \Big)\phi_{j\downarrow} \right\} \begin{pmatrix} \phi_{j\uparrow} \\ \phi_{j\downarrow} \end{pmatrix} \tag{4}$$

Having $\delta\phi$, we can now look at how we can formulate our density functional derivative. We need $\frac{\delta n}{\delta v_j}$, where δv_j is the derivative with respect to the j-th potential as defined by the descretization of the system. That means we have a j-length gradient of our density, with the differential density of each step being found as:

$$\frac{\delta n_i}{\delta v_j} = \frac{\delta \Psi_i}{\delta v_j}\Psi_i^\dagger + \Psi_i \frac{\delta \Psi_i^\dagger}{\delta v_j} \tag{5}$$

This leads to the differential density matrix δn.

$$\delta n = \begin{pmatrix} \delta\psi_\uparrow\psi_\uparrow^* + \psi_\uparrow\delta\psi_\uparrow^* & \delta\psi_\uparrow\psi_\downarrow^* + \psi_\uparrow\delta\psi_\downarrow^* \\ \delta\psi_\downarrow\psi_\uparrow^* + \psi_\downarrow\delta\psi_\uparrow^* & \delta\psi_\downarrow\psi_\downarrow^* + \psi_\downarrow\delta\psi_\downarrow^* \end{pmatrix} \tag{6}$$

Having established the potentials and the derivative method, we can formulate the 'density' vector that we will be calculating. Each potential has a 1-to-1 correspondence with with the various density and magnetization quantities.

$$\begin{pmatrix} n_a \\ n_b \\ m_{xa} \\ m_{xb} \\ m_{ya} \\ m_{yb} \\ m_{za} \\ m_{zb} \end{pmatrix} \longleftrightarrow \begin{pmatrix} V_a \\ V_b \\ B_{xa} \\ B_{xb} \\ B_{ya} \\ B_{yb} \\ B_{za} \\ B_{zb} \end{pmatrix} \tag{7}$$

We can define n_a and n_b from the spin density matrix that are generated as a function of the spinor wavefunctions. Explicitly, we can define the spin-density matrix below.

$$\begin{aligned} \underline{\underline{n}}(\mathbf{r}) &= \sum_i^N \Psi_i(\mathbf{r})\Psi_i^\dagger(\mathbf{r}) \\ &\equiv \begin{pmatrix} n_{\uparrow\uparrow}(\mathbf{r}) & n_{\uparrow\downarrow}(\mathbf{r}) \\ n_{\downarrow\uparrow}(\mathbf{r}) & n_{\downarrow\downarrow}(\mathbf{r}) \end{pmatrix} \end{aligned}$$

Having defined our spin-density array, we can calculate the total density $n(\mathbf{r}) \equiv m_0(\mathbf{r})$ where we can define the magnetization vector by using the spin-density matrix and the Pauli matrices.

$$m_i(\mathbf{r}) = \mathrm{Tr}\{\sigma_i \underline{\underline{n}}(\mathbf{r})\} \tag{8}$$

Here, i runs over the x, y, and z components, respectively. The relationship then allows us to calculate the magnetization from spin-density matrix calculated from our Hamiltonian.

$$\vec{m}(\mathbf{r}) = \begin{pmatrix} m_0(\mathbf{r}) \\ m_1(\mathbf{r}) \\ m_2(\mathbf{r}) \\ m_3(\mathbf{r}) \end{pmatrix} = \begin{pmatrix} n_{\uparrow\uparrow}(\mathbf{r}) + n_{\downarrow\downarrow}(\mathbf{r}) \\ n_{\uparrow\downarrow}(\mathbf{r}) + n_{\downarrow\uparrow}(\mathbf{r}) \\ i(n_{\uparrow\downarrow}(\mathbf{r}) - n_{\downarrow\uparrow}(\mathbf{r})) \\ n_{\uparrow\uparrow}(\mathbf{r}) - n_{\downarrow\downarrow}(\mathbf{r}) \end{pmatrix} \tag{9}$$

This means we now have a prescription to calculate the derivative with respect to δv_j for all measurable values.

$$\delta\vec{m}(\mathbf{r}) = \begin{pmatrix} (\delta\psi_\uparrow\psi_\uparrow^* + \psi_\uparrow\delta\psi_\uparrow^*) + (\delta\psi_\downarrow\psi_\downarrow^* + \psi_\downarrow\delta\psi_\downarrow^*) \\ (\delta\psi_\uparrow\psi_\downarrow^* + \psi_\uparrow\delta\psi_\downarrow^*) + (\delta\psi_\downarrow\psi_\uparrow^* + \psi_\downarrow\delta\psi_\uparrow^*) \\ i((\delta\psi_\uparrow\psi_\downarrow^* + \psi_\uparrow\delta\psi_\downarrow^*) - (\delta\psi_\downarrow\psi_\uparrow^* + \psi_\downarrow\delta\psi_\uparrow^*) \\ (\delta\psi_\uparrow\psi_\uparrow^* + \psi_\uparrow\delta\psi_\uparrow^*) - (\delta\psi_\downarrow\psi_\downarrow^* + \psi_\downarrow\delta\psi_\downarrow^*) \end{pmatrix} \tag{10}$$