Hyperfine-induced decoherence in triangular spin-cluster qubits

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We investigate hyperfine-induced decoherence in a triangular spin-cluster for different qubit encodings. Electrically controllable eigenstates of spin chirality ($C_z$) show decoherence times that approach millisecond, two orders of magnitude longer than those estimated for the eigenstates of the total spin projection ($S_z$) and of the partial spin sum ($S_{12}$). The robustness of chirality is due to its decoupling from both the total- and individual-spin components in the cluster. This results in a suppression of the effective interaction between $C_z$ and the nuclear spin bath.

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Introduction — Molecular nanomagnets represent a varied class of spin clusters, whose physical properties can be extensively engineered by chemical synthesis. This makes them candidate systems for the implementation of spin-cluster qubits. While most of the attention has been so far focused on the use of the total-spin projection ($S_z$) as a computational degree of freedom (DOF), it has been recently realized that alternative encodings would enable the use of electric - rather than magnetic - fields for the qubit manipulation. In particular, transitions between states of opposite spin chirality ($C_z = (4/\sqrt{3})s_1 \cdot s_2 \times s_3$) can be induced in antiferromagnetic triangles with Dzyaloshinskii-Moriya interaction. Spin-electric coupling constants compatible with ns gating times $\tau_g$ have been predicted by effective models and microscopic ab initio calculations. Further investigation is indeed required in order identify specific molecular nanomagnets with large spin-electric coupling, or to enhance such coupling by introducing suitable chemical substitutions in existing systems.

In order to assess the suitability of spin chirality for applications in quantum-information processing, its $\tau_g$ has to be contrasted with a characteristic decoherence time $\tau_d$. At low temperatures, quantum coherence in molecular nanomagnets is limited by the coupling to the nuclear spin environment, with typical values of $\tau_d$ in the microsecond range. All the existing literature is however concerned with linear superpositions of different $S_z$ eigenstates. Here we theoretically investigate the dependence of hyperfine-induced decoherence on the qubit encoding within a prototypical spin-cluster qubit, consisting of an antiferromagnetic spin triangle. In particular, we consider three different DOF, namely $S_z$, $C_z$, and the partial spin sum $S_{12}$ ($S_{12} = s_1 + s_2$), whose value - like that of $C_z$ - can be controlled through the spin-electric coupling. Since the optimal candidate system has not been identified yet, we refer here to a prototypical molecular spin-cluster qubit, with a typical electron-spin Hamiltonian and bath of nuclear spins. While the quantities of interest might to some extent vary from one molecular nanomagnet to another, the hyperfine-induced decoherence presents striking differences in the three considered DOF, that are not expected to depend on the specific features of the spin-cluster qubit.

Qubit encodings in the spin triangle — We consider a triangle of $s = 1/2$ spins, with dominant antiferromagnetic coupling and Zeeman interaction:

$$H_0 = J \sum_{i=1}^{3} s_i \cdot s_{i+1} + g \mu_B B \cdot S.$$  (1)

An additional term $H_1$ determines the expression of the lowest eigenstates $|0\rangle$ and $|1\rangle$, belonging to the ground state $S = 1/2$ quadruplet. As discussed in the following, the robustness of the spin-cluster qubit with respect to hyperfine-induced decoherence strongly depends on the distinguishability between $|0\rangle$ and $|1\rangle$ in terms not only of total spin orientation, but also of spin texture. Hereafter, we thus discuss these features in some detail in two relevant cases:

$$H_1^{C_z} = D z \cdot \sum_{i=1}^{3} s_i \times s_{i+1},$$  (2)

$$H_1^{S_{12}} = (J_{12} - J) s_1 \cdot s_2.$$  (3)
The term $H^C_1$ accounts for the Dzyaloshinskii-Moriya interaction in a spin triangle with, e.g., $D_{3h}$ symmetry [12]. For $H^c_1 = H_0 + H^C_1$, the four lowest eigenstates can be labelled after the value of the spin chirality $C_z = (4/\sqrt{3})S_z\times\mathbf{s}$, and the Dzyaloshinskii-Moriya term can be rephrased as: $H^C_1 = \Delta C_z S_z$, with $\Delta = D\sqrt{3}$ [2]. In particular, if the magnetic field is oriented parallel to the principal axis of the molecule (B $\parallel \hat{z}$), the eigenstates $|C_z, S_z\rangle$ read: $| \pm 1, \pm 1/2 \rangle = (| \uparrow \uparrow \rangle + e^{\pi i/2} | \uparrow \downarrow \rangle + e^{\pi i/3} | \downarrow \uparrow \rangle) / \sqrt{3}$ and $| \pm 1, -1/2 \rangle = \sigma_z^2 | \pm 1, \pm 1/2 \rangle$, where $\sigma_z^2$ is the Pauli operator acting on $s_z$. Both $S_z$ and $C_z$ commute with the electron-spin Hamiltonian $H_e$, which makes them suitable as computational DOF. In the first case, the logical states are:

$$|0\rangle_{S_z} = |S_z = -1/2; C_z = +1\rangle, \quad |1\rangle_{S_z} = |S_z = +1/2; C_z = +1\rangle.$$  

The expectation values of $s_z$ are oriented along the magnetic field, are identical for the three spins, and change sign with the qubit state [Fig. 1(a)]:

$$\langle 1 | s_z | 1 \rangle_{S_z} = -\langle 0 | s_z | 0 \rangle_{S_z} = 1/6.  \tag{4}$$

If the computational DOF is identified with spin chirality, the logical states are instead:

$$|0\rangle_{C_z} = |C_z = +1; S_z = -1/2\rangle, \quad |1\rangle_{C_z} = |C_z = -1; S_z = -1/2\rangle,$$

and the expectation values of the three spins are independent on the qubit state [Fig. 1(a)]:

$$\langle 1 | s_z | 1 \rangle_{C_z} = \langle 0 | s_z | 0 \rangle_{C_z} = -1/6.  \tag{5}$$

Such condition is however not general. In fact, if the applied magnetic field is tilted with respect to the z axis, $\mathbf{B} = B(\sin \theta \mathbf{x} + \cos \theta \mathbf{z})$, $C_z$ is still a good quantum number, but $\langle k | s_z | k \rangle_{C_z}^\theta$ (with $k = 0, 1$) are always oriented along $\mathbf{B}'_k = (B_x, 0, B_z \pm \Delta / g\mu_B)$. Eigenstates of opposite chirality are thus characterized by different orientations of the spin expectation values [see Fig. 1(b)]:

$$\langle k | s_z | k \rangle_{C_z}^\theta = \sin \alpha_k / 6, \quad \langle k | s_z | k \rangle_{C_z}^\theta = \cos \alpha_k / 6,  \tag{6}$$

where $\alpha_k = \arctan \left( -\frac{\Delta}{B\cos \theta} \right)$ and $\chi = \pm 1$ for $\Delta \geq Bg\mu_B$.

If no Dzyaloshinskii-Moriya interaction is present and one exchange coupling differs from the other two, the term $H^C_1$ is replaced by $H^C_2$ (Eq. 3). For $H^C = H_0 + H^C_2$, the four lowest eigenstates can be labelled after the partial sum of the first two spins, rather than the spin chirality: $|S_{12}, S_z\rangle$, where $S_{12} = 0, 1$. Their expressions read: $|0, +1/2\rangle = (| \uparrow \uparrow \rangle - | \downarrow \downarrow \rangle) / \sqrt{2}$, $|1, +1/2\rangle = (| \uparrow \downarrow \rangle + | \downarrow \uparrow \rangle) / \sqrt{2}$, and $|S_{12}, -1/2\rangle = \sigma_z^2 | \pm 1, \pm 1/2 \rangle$. Choosing $S_{12}$ as the computational DOF, one has:

$$|0\rangle_{S_{12}} = |S_{12} = 0; S_z = -1/2\rangle, \quad |1\rangle_{S_{12}} = |S_{12} = 1; S_z = -1/2\rangle.$$  

As far as the spin expectation values are concerned, $S_{12}$ represents an intermediate case between $S_z$ and $C_z$. The qubit states have in fact identical values for the total spin, $\langle 0 | \mathbf{S} | 0 \rangle_{S_{12}} = \langle 1 | \mathbf{S} | 1 \rangle_{S_{12}}$, like $C_z$, but they strongly differ in terms of spin texture, like $S_z$ [Fig. 1(a)]:

$$\langle 0 | s_z, i = 1, 2 | 0 \rangle_{S_{12}} = 0, \quad \langle 0 | s_z, i = 3 | 0 \rangle_{S_{12}} = -1/2  \tag{7a}$$

$$\langle 1 | s_z, i = 1, 2 | 1 \rangle_{S_{12}} = -1/3, \quad \langle 1 | s_z, i = 3 | 1 \rangle_{S_{12}} = 1/6.  \tag{7b}$$

**Nuclear spin and hyperfine interactions** — The decoherence of the spin-cluster qubit is investigated by simulating the coupled dynamics of electron and nuclear spins, induced by the Hamiltonian $H = H_e + H_n + H_{en}$. The qubit and the nuclear environment are initialized respectively in the linear superposition $| \psi(0) \rangle = | 0 \rangle + | 1 \rangle$ and the mixed state $| \rho \rangle = \sum_i | I_i \rangle | I_i \rangle$ and $m_i^z$ are the projections along the magnetic field direction of the $N_n$ nuclear spins. In the pure-dephasing regime, each state $| \Psi_I(0) \rangle = \frac{1}{\sqrt{2}} | 0 \rangle + | 1 \rangle$ evolves into: $| \Psi_I(t) \rangle = \frac{1}{\sqrt{2}} | 0 \rangle | 0 \rangle + | 1 \rangle | 1 \rangle$, where $| 0 \rangle$ (| 1 \rangle) can be regarded as the state of the nuclear bath conditioned upon the qubit being in the | 0 \rangle (| 1 \rangle) state. The degree of decoherence in the reduced density matrix of the qubit, $p_{en} = Tr_n \{ \sum_I P_I | \Psi_I(t) \rangle \langle \Psi_I(t) | \}$, is given by the so-called decoherence factor: $r_{\perp} = Tr \{ \sum_I P_{I\perp} | r_{\perp} \}$. The nuclear spin bath we consider consists of $N_n = 200$ hydrogens ($I = 1/2$), whose positions $r^\alpha_i$ are randomly generated so as to reproduce typical values of the spin density and the electron-nuclear distances $d_{en} = | r_i^e - r^\alpha_i |$, where $r_i^e$ are the positions of electron spins [Fig. 1(c)] [13]. The nuclear-spin Hamiltonian $H_n$ includes Zeeman and dipole-dipole terms: $H_n = B \sum_i \omega_i | I_p \rangle \langle I_p | - 3 | I_p \rangle \langle I_p | r_{pp_i} | r_{pp_i} \rangle / | r_{pp_i} |$, where $D_{en} = (\mu_0 / 4\pi) \mu_i^2 \gamma_i^2$ and $r_{pp_i} = r_i^e - r_i^\alpha$. Electron and nuclear spins are coupled by dipole-dipole and contact interactions: $H_{en} = D_{en} \sum_i \sum_{j<i} | s_i \rangle \langle I_p | - 3 | s_i \rangle \langle r_{pp_i} | r_{pp_i} \rangle$. 

**FIG. 2** (color online) Time dependence of the decoherence factor $r$ for the three qubit encodings: $S_z$ (black), $S_{12}$ (red), and $C_z$ (green for $\theta = 0$ and blue for $\theta = \pi / 8$). The curves are averaged over $N = 5 \times 10^3$ randomly generated initial states $| 2 \rangle$ of the nuclear bath. Inset: Statistical distribution (squares) of the parameter $\delta$, and corresponding Gaussian fits (solid lines); same convention as above for the colors.
f_{ip}]/r^3_{ip} + \sum_i \alpha_i \mathbf{s}_i \cdot \mathbf{I}(i)$, where $D_{en} = (\mu_0/4\pi)\mu_n \mu_e \gamma_e \gamma_n$ and $r_{ip} = r_i - r_p$. The contact terms $\alpha_i$, whose effect will be considered in the final part of the paper, couples electron and nuclear spins belonging to the same magnetic center.

The dephasing arises from the qubit-state dependent dynamics of the nuclear bath, generated by an effective Hamiltonian $\mathcal{H}$. We derive $\mathcal{H}$ from the above specified $H = H_e + H_n + H_{en}$ in a two-step procedure [13][10]. We first project the single-electron spin operators $\sigma_{\alpha,i}$ onto the $S = 1/2$ quadruplet: $\mathcal{P}_{S=1/2} = \sum_{i,j=0}^{\pm 1/2} \langle |i\rangle \langle j| \sigma_{ij} \rangle$, where $\sigma_{ij} = |i\rangle \langle j|$ and $|i\rangle$ are the eigenstates of $\sigma_i$. We then apply a Schrieffer-Wolff transformation, that removes from the Hamiltonian the terms that are off-diagonal in the basis of electron-spin eigenstates $|i\rangle$ [14][15][16], and finally neglect energy non-conserving terms (secular approximation). The resulting Hamiltonian reads: $\mathcal{H} = \sum_{k=0,1} |k\rangle \langle k| \otimes \mathcal{H}_k$, where

$$\mathcal{H}_k = \sum_{p=1}^{N_n} \omega_p \mathbf{I}_p + \sum_{p \neq q} \left( A^{pq}_i \mathbf{I}_p \cdot \mathbf{I}_q + B^{pq}_i \mathbf{I}_p \times \mathbf{I}_q \right) \quad (8)$$

and $\mathbf{z}' \equiv B/B$. Differences between $\mathcal{H}_0$ and $\mathcal{H}_1$ result from the hyperfine interactions, and are responsible for the qubit decoherence, being $\tau(t) = \langle \mathcal{I} \rangle \exp[i\mathcal{H}_k t/\hbar] \exp[-i\mathcal{H}_0 t/\hbar] / \langle \mathcal{I} \rangle$. In particular, the quantities $(\omega_0^0 - \omega_0^j)$ are linear in $\mathcal{H}_n$, and essentially result from differences in the magnetic field induced by the nuclear spin $\mathbf{l}_k$ at the electron-spin positions $\mathbf{r}_i$ (see below). The terms $(A^{pq}_i - A^{pq}_j)$ and $(B^{pq}_i - B^{pq}_j)$ are instead quadratic in $\mathcal{H}_n$, and result from the qubit-state dependence of the couplings between pairs of nuclei, mediated by virtual transitions of the electron spins. The time evolution of the nuclear states $|\mathcal{I}_k\rangle$ is computed within the pair-correlation approximation, where the nuclear dynamics is traced back to independent flip-flop transitions between pairs of nuclear spins [17][19].

**Hyperfine-induced decoherence** — The fastest contribution to dephasing in the spin-cluster qubit is related to inhomogeneous broadening, and typically takes place on time scales that are much shorter than those characterizing the dynamics of the nuclear bath ($\tau_n \sim \hbar/|B^{k}_{n,m}| \sim 10^2 \mu$s). Such contribution results from the renormalization of the energy gap between the states $|0\rangle$ and $|1\rangle$ induced by the hyperfine interactions: $\delta_\tau = \sum_{k=0,1} \langle -1 \rangle^k (|k\rangle \mathcal{I}|k\rangle) \simeq \sum_{\alpha \beta} (\omega_0^\alpha - \omega_0^\beta) m_\tau$. Being the nuclear spin bath initially in a mixture of states $|\mathcal{I}\rangle$, the decoherence factor evolves as: $r(t \ll \tau_n) \simeq e^{-i |B_n-E_k|} \sum_\mathcal{I} P_\mathcal{I} e^{-i \delta_\tau t}$, while $|\mathcal{I}(t \ll \tau_n) \rangle \simeq |\mathcal{I}\rangle$. In first order in $\mathcal{H}_n$, $\delta_\tau$ can be regarded as a function of the Overhauser field at the electron-spin site: $\delta_\tau \simeq \mu_B g \sum_{i} B^{ij}_{hf}(\mathbf{r}_i') \cdot \left[ |\langle s_i|0\rangle - |\langle s_i|1\rangle| \right]$, (9)

where $B^{ij}_{hf}(\mathbf{r}_i') = D_{en} \sum_m m_p z' \left[ 3(z' \cdot \mathbf{r}_{ip}) \mathbf{r}_{ip} / r_{ip}^3 \right]$. In the case of the $S_z$ qubit (see Eq. [4]), $\delta_\tau \simeq -\mu_B g / 3 \sum_i B^{ij}_{hf,z'}(\mathbf{r}_i')$. The statistical distribution $N(\delta_\tau)$ is reported in the inset of Fig. 2 (black squares) for $5 \times 10^4$ initial nuclear states $|\mathcal{I}\rangle$, randomly generated from a flat probability distribution $P_\mathcal{I} = 1/2^{N_n}$.

$N(\delta_\tau)$ is well fitted by a Gaussian function (solid line), with $\sigma_{S_z} = 9.0$ neV. Correspondingly, the decay of $\langle |r(t)| \rangle$ (black line in Fig. 2) is approximately Gaussian, and its characteristic time scale is $10^2$ ns. In the case of the $S_{12}$ qubit, the three electron spins are no longer equivalent: $\delta_{12} \simeq -\mu_B g / 3 \left[ 2B^{ij}_{hf,z'}(\mathbf{r}_i') - B^{ij}_{hf,x}(\mathbf{r}_i') - B^{ij}_{hf,z}(\mathbf{r}_i') \right]$. However, the statistical distribution of $\delta_{12}$ strongly resembles that of $S_z$ (see the red squares in the figure inset, and the Gaussian fit with $\sigma_{S_{12}} = 9.4$ neV), and so does the time evolution of the decoherence factor (red curve in the main panel). In fact, since the distances $d_{ee}$ between electron spins are larger than the smallest $d_{en}$ [see Fig. 1(c)] [14], the spatial fluctuations of the Overhauser field within the spin triangle are comparable to its average value. In spin clusters with larger $d_{en}/d_{ee}$ ratios (not shown here), spatial fluctuations of $B_{nf}(\mathbf{r})$ are relatively small. As a result, $\delta_{12} \ll \delta_{S_z}$, and the $S_{12}$ qubit is less affected by inhomogeneous broadening than $S_z$.

In the case of the $C_z$ qubit and for $B \parallel \mathbf{z}$, the Overhauser field does not renormalize the energy difference between the states $|0\rangle$ and $|1\rangle$, that have identical expectation values for all single-spin projections (Eqs. 5 [9]). The leading contribution to $\delta_{C_z}$ is given by terms that are second order in the hyperfine Hamiltonian, $\delta_{C_z} = \frac{\mu_B g}{3} \sum_{i} m_i \hat{m}_i$, and its modulus is here $5 \times 10^4$ orders of magnitude smaller than that of $\delta_{S_z}$ and $\delta_{S_{12}}$. Correspondingly, no inhomogeneous broadening occurs in the considered time scale (green curve). For a tilted magnetic field ($\theta \neq 0$), states of opposite chirality have different expectation values $\langle s_i \rangle$ (see Eq. 6), and thus couple differently to the Overhauser field. The leading contribution to the renormalization of the energy difference reads: $\delta_{C_z} \simeq (\mu_B g)/3 \sum_{i} m_i \left[ \sin \alpha_{\mathbf{B}_{hf,z'}} + \cos \alpha_{\mathbf{B}_{hf,z'}} \right]$, where $\mathbf{x'} \perp \mathbf{z'}$ and lies in the $xz$ plane. The statistical distribution of $\delta_{C_z} (\theta = \pi/8)$ and the resulting...
qubit dephasing are reported in Fig. 2 (\(\sigma_{C_z} = 4.5\) neV, blue curve).

Nuclear spin evolution in the field of electron spins creates entanglement in the initially separable state \(|\Psi_I(0)\rangle\), and causes additional decoherence on the time scale of nuclear dynamics, \(\tau_d\). In order to single out this contribution, we compute the function: \(r_m(t) = \sum_{2} P_{2}\langle r_{2}(t)\rangle\). In the case of the S\(_2\) qubit, electron-nuclear correlations result in a decay of \(r_m\) in the \(\mu s\) time scale (Fig. 4 black curve). The decay is induced by the interplay of the dipolar interactions between the nuclei and of the term \(\sum_{p}(\omega_{p}^{0} - \omega_{p}^{z})I_{p}^{z}\), whose expectation value gives \(\delta_{I}^{z}\). A similar time dependence for \(r_m\) is obtained in the case of the S\(_{12}\) qubit (red curve). Here, the same terms in the effective Hamiltonian \(H\) dominate, and have similar expectation values: \(\delta_{I}^{S_{z}} \simeq \delta_{I}^{S_{12}}\) (see the inset of Fig. 2). This quantity (\(\delta_{I}^{S_{z}}\)) is about 5 orders of magnitude smaller for the C\(_z\) qubit, if \(B \parallel \hat{z}\). As a result, the dynamics of the nuclear bath is largely independent on the qubit state in the considered time range, and no appreciable decoherence takes place (green curve). This is no longer the case if the magnetic field is not aligned with the principal axis of the spin triangle: in fact, the decoherence time rapidly decreases as \(\delta_{I}^{C_{z}}(\theta)\) increases with the tilting angle \(\theta\) (blue curves).

We finally investigate the possible contribution to decoherence of the contact terms. Such contribution is qualitatively different from that of the dipolar interactions, for it results from the relatively strong coupling with the electron spins of few \((N^e_n \sim N_e \ll N_n)\) nuclei. Here, the \(N^e_n = N_e = 3\) additional nuclear spins are localized at the electron spin sites \(r^e_i\) and are assumed for simplicity identical to the remaining \(N^d_n = N_n - N^e_n = 200\) nuclei. The inequivalence between the \(N^e_n\) and \(N^d_n\) nuclear spins, resulting from strong coupling of the former ones with the electron spins, warrants the factorization of the decoherence factor: \(r(t) = r_{c}(t) r_{d}(t)\). The time evolution of \(r_{c}(t)\) is reported in Fig. 4 for \(a_p = 1\) mK and \(a_p = 10\) mK [panels (a) and (b), respectively]. In the case of \(S_z\) (black curve) and \(S_{12}\) (red), \(r_c\) is responsible for the fast oscillations, while the decay is due to \(r_d\) (dotted lines). Oscillations of the decoherence factor caused by the contact interactions are also present in the case of C\(_z\) qubit (figure insets). These take place on a time scale which is much longer than that of \(S_z\) and \(S_{12}\), but much shorter than the one that characterizes the contribution to the decoherence of \(C_z\) due to dipolar coupling. The chirality qubit also presents a different dependence on the contact coupling constant \(a_i\) with respect to \(S_z\) and \(S_{12}\). A comparison between the two panels shows in fact that the characteristic time scale of the oscillations in \(r_c\) is \(\tau_{d} \sim h/a_p\) for \(S_z\) and \(S_{12}\), and \(\tau_{d} \sim h\delta_{ij}/a^2_{ij}\) for \(C_z\), where \(\delta_{ij} \sim \min\{\Delta, \mu g \mu_B B\}\) is the smallest difference between eigenvalues of \(H_z\). Like in the case of dipolar interactions discussed above, the leading contributions of contact interaction to \(H\) are thus quadratic in the hyperfine Hamiltonian for \(C_z\), and linear for the other two DOF.

In conclusion, we have shown that the nuclear-induced decoherence in a prototypical spin triangle strongly depends on the qubit encoding. In particular, the decoherence time of the chirality qubit approaches the ms range, i.e. several orders of magnitude larger than the gating times predicted for the manipulation by means of electric fields. Such robustness results from the decoupling of \(C_z\) from both the total spin orientation and the spin texture, preserved by the alignment of the magnetic fields in the direction normal to the spin triangle. The eigenstates of \(S_{12}\) are characterized instead by different spin textures, and thus couple differently to the nuclear spin bath. This results in decoherence times comparable to those of the total spin projection \(S_z\). Larger decoherence times for \(S_{12}\) could be obtained in spin clusters where the distance between electron spins is much smaller than that between electron and nuclear spins. Finally, the presence of a strong contact interaction between nuclear and electron spins introduces an oscillating behavior in the decoherence factor of all the DOF, with characteristic time scales that are \(10^2\) times larger for spin chirality than for \(S_z\) and \(S_{12}\).

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[20] The time evolution of $r_{m}$ obtained for $S_{z}$ with the full Hamiltonian $\mathcal{H}$ is in fact indistinguishable from that obtained by keeping in $\mathcal{H}$ only the terms that are linear in the hyperfine couplings.
[21] The hyperfine field induced by the contact terms is in fact of the order of $1 \div 10 \text{T}$. This makes the occurrence of a flip-flop transition between an $I_{p}$ with an $I_{q}$ without contact interaction highly unlikely, being: $|\omega_{p} - \omega_{q}| \sim a_{p} \gg B_{pq} \sim D_{nn}/r_{pq}^{3}$. 