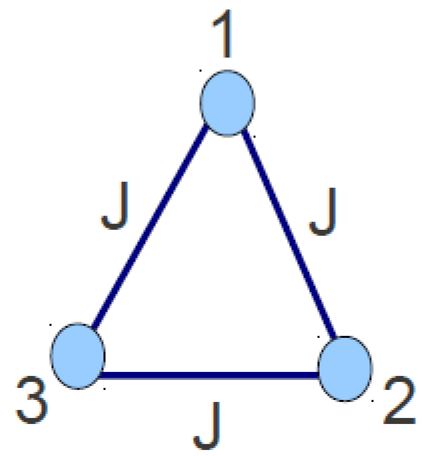
Quantum Discord in Ground and Thermal States of Molecular Magnets

Indrani Bose
Department of Physics
Bose Institute

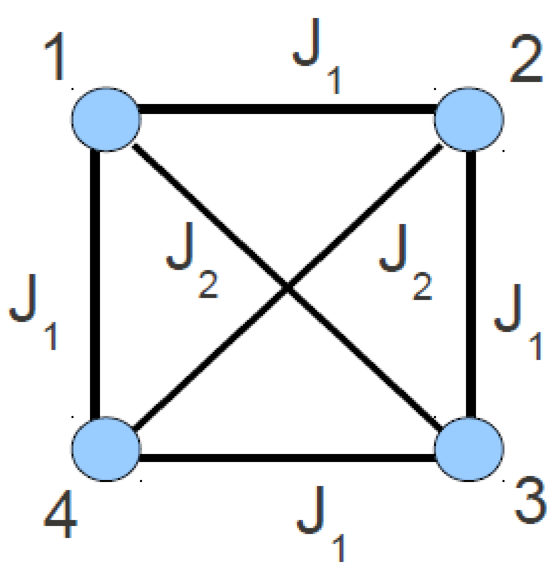
A number of molecular magnets are represented by weakly-interacting small spin clusters

(Haraldsen et al., PRB 71 064403 (2005), Bose and Tribedi, PRA 72 022314 (2005))

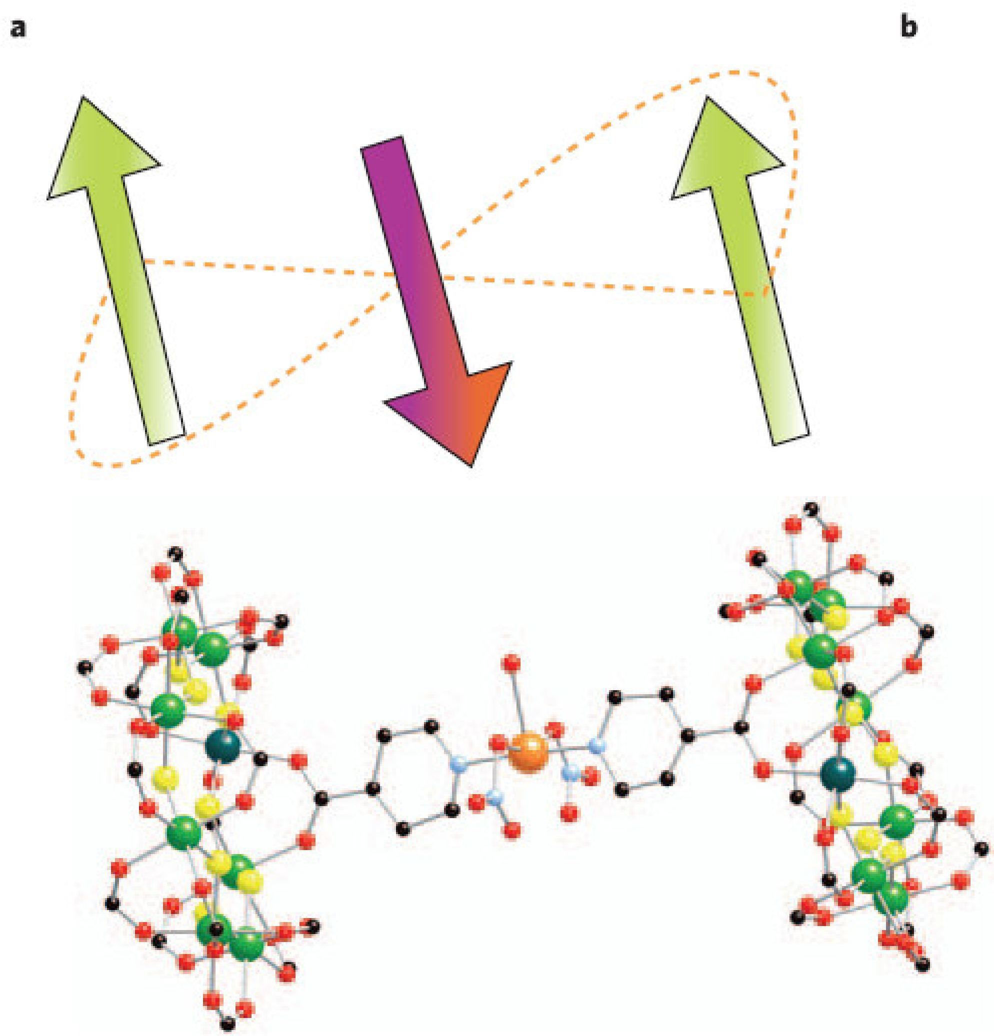
Spin Trimer



Spin Tetramer



A recently engineered molecular complex Cr_7Ni-Cu^{2+}-Cr_7Ni serves as a three-qubit system (Timco et al. Nature Nanotechnology 4 173 (2009))



Suggestion: microwave pulse sequences can be used to generate maximally entangled states in such molecules

Our objective: to quantify quantum correlations in the ground and thermal states of spin trimers and tetramers in terms of a new measure, quantum discord (QD), different from entanglement

Certain separable mixed states have zero entanglement but non-zero discord

Quantum mutual information $I(\rho_{AB})$: measures total correlations (quantum + classical) in a bipartite quantum system

$$I(\rho_{AB}) = S(\rho_A) + S(\rho_B) - S(\rho_{AB})$$

$\rho_{A(B)}$: reduced density matrix of subsystem A (B)

ρ_{AB}: density matrix of total system

$S(\rho) = - \mathrm{Tr}(\rho \log_2 \rho)$ is von Neumann entropy

Quantum discord (QD):

$Q(\rho_{AB}) = I(\rho_{AB}) - C(\rho_{AB})$

$C(\rho_{AB})$ measures classical correlations

$Q(\rho_{AB})$ measures quantum correlations

How does one quantify $Q(\rho_{AB})$?

Classical information theory

Shannon entropy quantifies uncertainty about random variable A before we learn its value

$$H(A) = -\sum_a p(a) \log p(a)$$

Correlation between two random variables A, B, measured by mutual information

$$I(A, B) = H(A) + H(B) - H(A, B)$$

Alternative definition:

$$J(A, B) = H(A) - H(A \mid B)$$

H (A | B): conditional entropy, quantifies uncertainty about A given knowledge of B

$$H(A, B) = -\sum_{a,b} p(a,b) \log p(a,b)$$

$$H(A \mid B) = -\sum_{a,b} p(a, b) \log p(a \mid b)$$

Since p(a, b) = p (b) p (a | b)

$$H(A, B) = -\sum_{a,b} p(a, b) \log \{ p(b)\, p(a \mid b) \}$$

$$H(A,B) = -\sum_{a} p(b) \log p(b)$$

$$-\sum_{a,b} p(a, b) \log p(a \mid b)$$

$$= H(B) + H(A \mid B)$$

Thus,

$I(A, B) = J(A, B)$

Quantum Information Theory:

Consider a bipartite system with parties A and B

Quantum Mutual Information:

$$I(\rho_{AB}) = S(\rho_A) + S(\rho_B) - S(\rho_{AB})$$

$$J(\rho_{AB}) = S(\rho_A) - S(\rho_A \mid \rho_B)$$

$S(\rho_A) = -\,\mathrm{tr}\, \rho_A \log \rho_A$ (von Neumann entropy)

Are $I(\rho_{AB})$ and $J(\rho_{AB})$ identical?

Quantum generalization of classical mutual information:

Replace

Classical probability distributions by density matrices

Shannon entropy by von Neumann entropy

What about $J(\rho_{AB})$?

Quantum conditional entropy $S(\rho_A | \rho_B)$

depends on type of measurement

Different measurement choices yield different results, so

$I(\rho_{AB}) \neq J(\rho_{AB})$

von Neumann measurement for party B can be written as

$B_k = V \Pi_k V^+$, $k = 0, 1$

$\Pi_k = |k\rangle\langle k|$

where V is a unitary operator with unit determinant

Projectors B_k: represent an arbitrary local measurement on B

Parametrize V as

$$V = \begin{pmatrix} \cos\frac{\theta}{2} & \sin\frac{\theta}{2}\exp(-i\phi) \\ \sin\frac{\theta}{2}\exp(i\phi) & -\cos\frac{\theta}{2} \end{pmatrix}$$

θ, φ: azimuthal and polar angles of a qubit over the Bloch sphere

We consider Von Neumann-type measurements on B defined in terms of a complete set of orthogonal projectors $\{\Pi_i\}$, corresponding to the set of possible outcomes i [2].

After measurement,

$$\rho_i = (I \otimes \Pi_i^B)\rho_{AB}(I \otimes \Pi_i^B)/p_i$$

where $p_i = tr((I \otimes \Pi_i^B)\rho_{AB}(I \otimes \Pi_i^B))$

$$S(\rho_{AB}|\{\Pi_i^B\}) = \sum_i p_i S(\rho_i)$$

Quantum Conditional Entropy: Alternative Definition [2]

$$J(\rho_{AB}|\{\Pi_i^B\}) = S(\rho_A) - S(\rho_{AB}|\{\Pi_i^B\})$$

Quantum Mutual Information : Alternative definition [2]

$$C(\rho_{AB}) = \max_{\{\Pi_i^B\}}(J(\rho_{AB}|\{\Pi_i^B\}))$$

A measure of **classical correlation** [2,3]

The difference between the total correlations $I(\rho_{AB})$ and the classical correlations $C(\rho_{AB})$ defines the **QD** [2]

$$\boldsymbol{Q(\rho_{AB}) = I(\rho_{AB}) - C(\rho_{AB})}$$

QD can be quantified analytically/numerically for two-qubit states

For spin Hamiltonians with specific features the two-spin reduced density matrix has the form

$$\rho_{ij} = \begin{pmatrix} a & 0 & 0 & f \\ 0 & b_1 & z & 0 \\ 0 & z & b_2 & 0 \\ f & 0 & 0 & d \end{pmatrix}$$

The eigenvalues of ρ_{ij} are:

$$\lambda_0 = \frac{1}{4}\left\{(1+c_3) + \sqrt{(c_4+c_5)^2 + (c_1-c_2)^2}\right\}$$

$$\lambda_1 = \frac{1}{4}\left\{(1+c_3) - \sqrt{(c_4+c_5)^2 + (c_1-c_2)^2}\right\}$$

$$\lambda_2 = \frac{1}{4}\left\{(1-c_3) + \sqrt{(c_4-c_5)^2 + (c_1+c_2)^2}\right\}$$

$$\lambda_3 = \frac{1}{4}\left\{(1-c_3) + \sqrt{(c_4-c_5)^2 + (c_1+c_2)^2}\right\}$$

with

$$c_1 = 2z + 2f$$

$$c_2 = 2z - 2f$$

$$c_3 = a + d - b_1 - b_2$$

$$c_4 = a - d - b_1 + b_2$$

$$c_5 = a - d + b_1 - b_2$$

Mutual information:

$$I\left(\rho_{AB}\right)=S\left(\rho_{A}\right)+S\left(\rho_{B}\right)+\sum_{\alpha=0}^{3}\lambda_{\alpha}\log_{2}\lambda_{\alpha}$$

where

$$\begin{aligned}S\left(\rho_{A}\right) &= -\frac{\left(1+c_{5}\right)}{2}\log_{2}\frac{\left(1+c_{5}\right)}{2}-\frac{\left(1-c_{5}\right)}{2}\log_{2}\frac{\left(1-c_{5}\right)}{2}\\ S\left(\rho_{B}\right) &= -\frac{\left(1+c_{4}\right)}{2}\log_{2}\frac{\left(1+c_{4}\right)}{2}-\frac{\left(1-c_{4}\right)}{2}\log_{2}\frac{\left(1-c_{4}\right)}{2}\end{aligned}$$

$$\rho_{ij}=\begin{pmatrix} a & 0 & 0 & f\\ 0 & b_{1} & z & 0\\ 0 & z & b_{2} & 0\\ f & 0 & 0 & d\end{pmatrix}$$

When $f = 0$, $a = d$ and $b_1 = b_2$, the maximization procedure for calculating the classical correlations can be carried out analytically

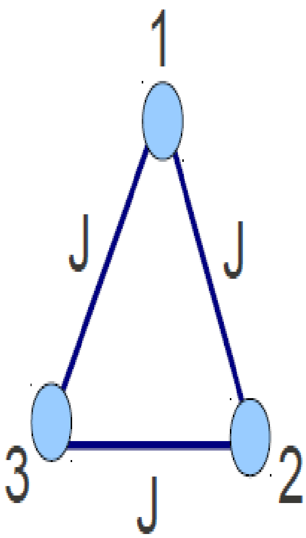
$$C\left(\rho_{AB}\right)=\frac{\left(1-c\right)}{2}\log_{2}(1-c)+\frac{\left(1+c\right)}{2}\log_{2}(1+c)$$

where

$$c = \max\left(\left|c_1\right|, \left|c_2\right|, \left|c_3\right|\right)$$

$$\begin{aligned}
Q(\rho_{AB}) &= I(\rho_{AB}) - C(\rho_{AB}) \\
&= \frac{1}{4}\big[(1-c_1-c_2-c_3)\log_2(1-c_1-c_2-c_3) + (1-c_1+c_2+c_3)\log_2(1-c_1+c_2+c_3) \\
&\quad + (1+c_1-c_2+c_3)\log_2(1+c_1-c_2+c_3) + (1+c_1+c_2-c_3)\log_2(1+c_1+c_2-c_3)\big] \\
&\quad - \frac{(1-c)}{2}\log_2(1-c) - \frac{(1+c)}{2}\log_2(1+c) \qquad (17)
\end{aligned}$$

Spin Trimer



The Hamiltonian describing the trimer of spin-1/2

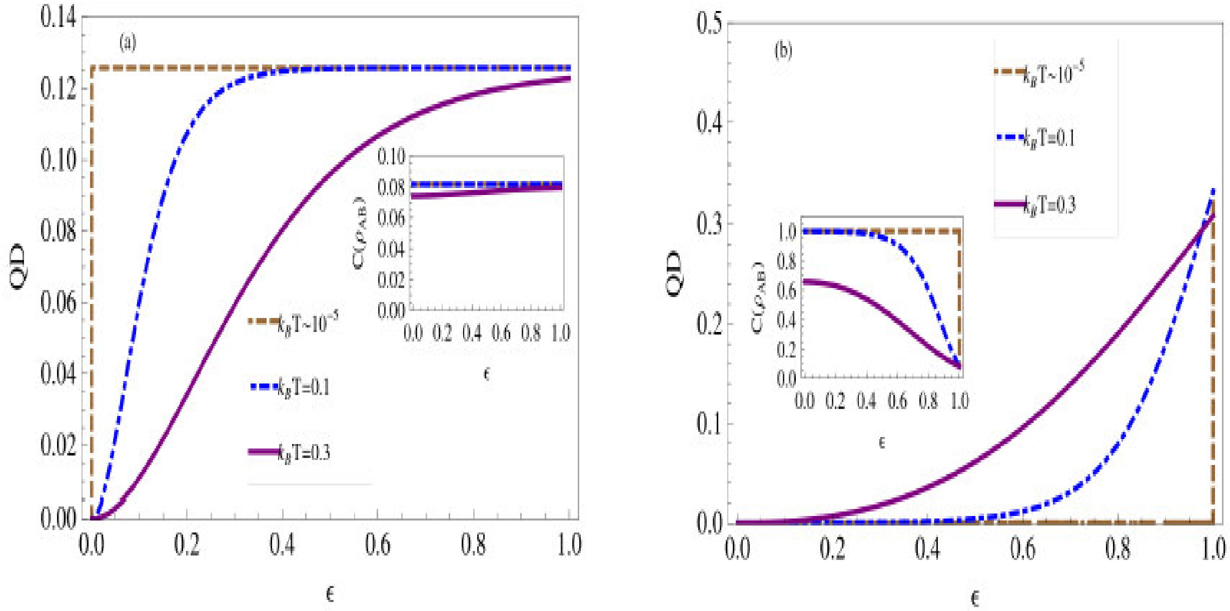
$$\boxed{H = J\sum_{i=1}^{3} S_i^z S_{i+1}^z + \varepsilon J \sum_{i=1}^{3} \left(S_i^x S_{i+1}^x + S_i^y S_{i+1}^y\right)}$$

In the **AFM** region J > 0, the value of the ground state QD is 0.125 whereas entanglement, as measured by concurrence, is zero in this case.

In the **FM** region J < 0, the value of the ground state QD is 0.333 and the concurrence is zero.

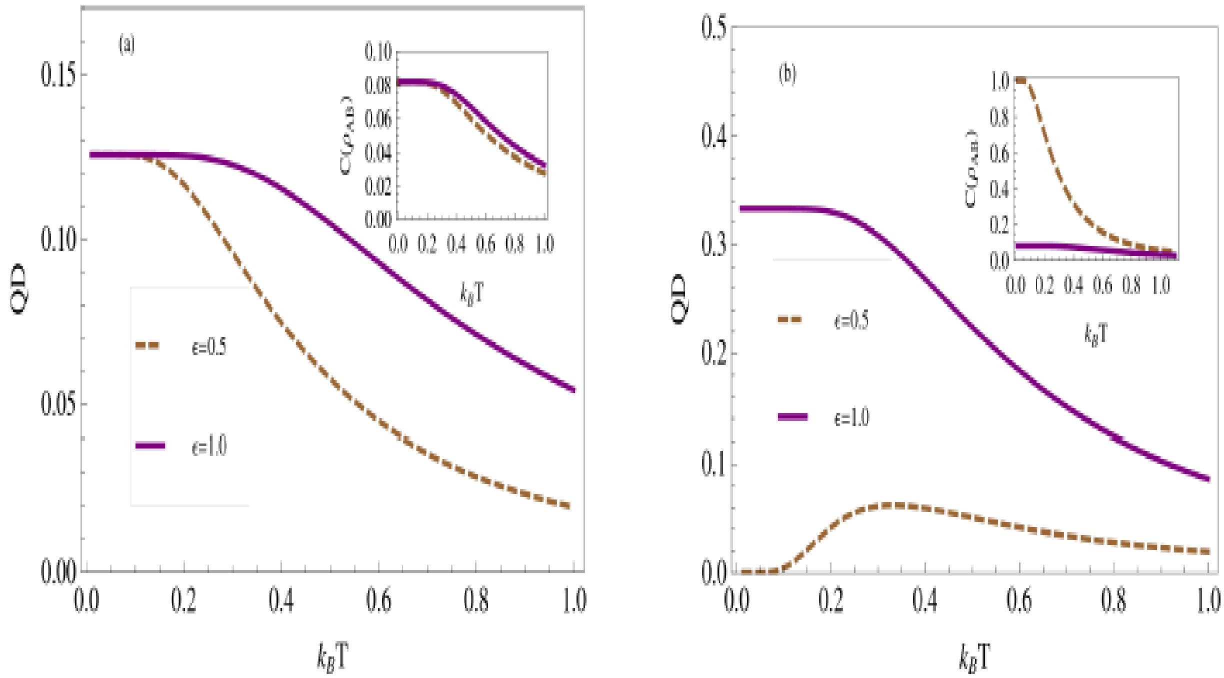
- QD has a lower value in the AFM region than the FM region
- In both the cases, value of $C(\rho_{AB})$ is 0.082

(Amit Kumar Pal, Indrani Bose, J. Phys. B (2011)



Variation of quantum discord (QD) and classical correlation $C(\rho_{AB})$ (inset) as a function of the anisotropy parameter ε for different temperatures in the (a) AFM and (b) FM cases with $|J| = 1$.

- In both the AFM and the FM cases, the QD increases with ε.
- At a fixed value of ε, the QD decreases with increasing T in the AFM region though non-monotonic behaviour is observed in the FM case.
- $C(\rho_{AB})$ increases slowly with ε for a fixed value of T in the AFM region whereas in FM case, it decreases with ε.

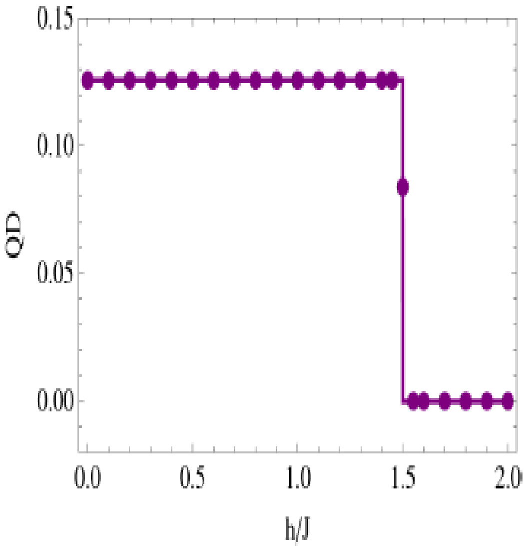


Variation of quantum discord (QD) and classical correlation $C(\rho_{AB})$ (inset) as a function of temperature T for different values of the anisotropy parameter ε in the (a) AFM and (b) FM cases with $|J| = 1$.

- QD goes to zero asymptotically with temperature in both the AFM and the FM cases.
- In the FM case, for low value of ε, the QD first increases with T, reaches a maximum value and then decreases. For high value of ε, the variation is similar to that in the AFM case.

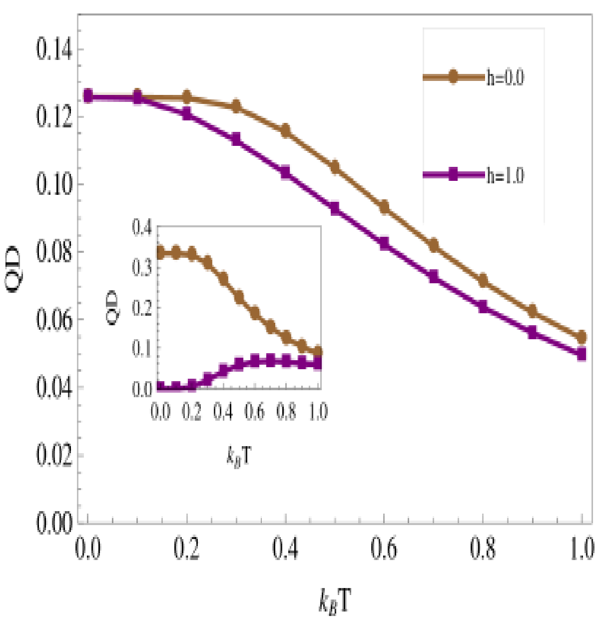
Spin Trimer With External Magnetic Field

Now we introduce an external magnetic field in the +z direction.



Variation of quantum discord (QD) with magnetic field h at T = 0. The QD remains constant in the range $0 \le h \le 3J/2$. At $h = 3J/2$, the value of QD is 0.0838764. When h is > $3J/2$, the QD is zero.

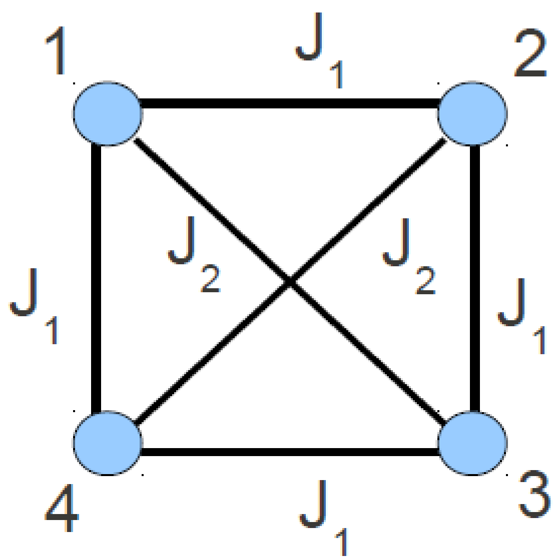
☼ Discontinuous jump of the QD at 1st order QPT point



Variation of quantum discord (QD) as a function of temperature for different values of h and ($|J|$ = 1.0) in the AFM and FM (inset) cases.

☼ The QD decays asymptotically with temperature.

Spin Tetramer



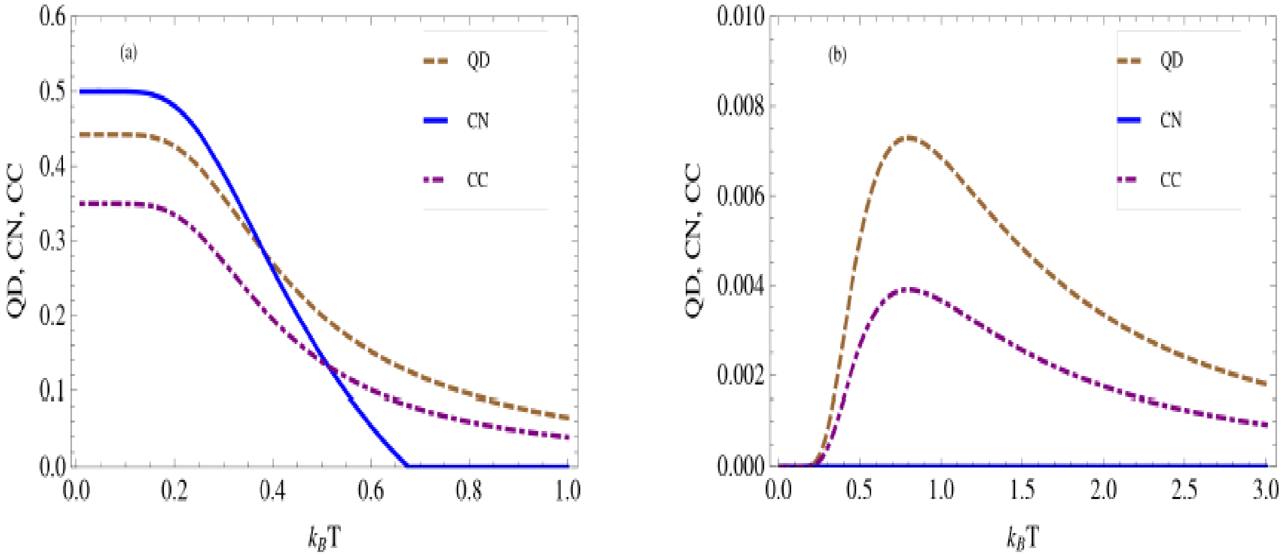
The Hamiltonian describing the tetramer of spin-1/2

$$\boxed{H = J_1 \sum_{i=1}^{4} \vec{S}_i . \vec{S}_{i+1} + J_2 \sum_{i=1}^{2} \vec{S}_i . \vec{S}_{i+2}}$$

At $J_1 = J_2$, a first order QPT separates two RVB ground states

QD has a discontinuity at the transition point

(Bose and Chattopadhyay PRA 66 062320 (2002), Wu et al., PRL 93 250404 (2004))



Variation of n.n. quantum discord (QD), concurrence (CN) and classical correlation (CC) as functions of temperature for (a) $J_1 > J_2$ ($J_1 = 2J_2 = 1.0$) and (b) $J_1 < J_2$ ($2J_1 = J_2 = 1.0$).

- When $J_1 > J_2$, both the CN and QD decreases with temperature but QD has non-zero values at temperature much higher than the value at which CN goes to zero.
- When $J_1 < J_2$, CN has zero value at all temperatures. The QD is zero at T=0, then it increases with T to reach a maximum value after which it decreases with temperature.

Quantum Correlation under Decoherence

Interaction of a quantum system with its environment $\longrightarrow$ **DECOHERENCE**

We consider interaction of qubits with their local environments.

In the Kraus Operator representation [6]

$$\boxed{\rho(t)=\sum_{\mu,\nu} E_{\mu,\nu}\rho(0)E_{\mu,\nu}^{\dagger}}$$

where $E_{\mu,\nu}=E_{\mu}\otimes E_{\nu}$ Is the Kraus operator

For a Dephasing channel

$$|0\rangle_S|0\rangle_E \longrightarrow |0\rangle_S|0\rangle_E$$

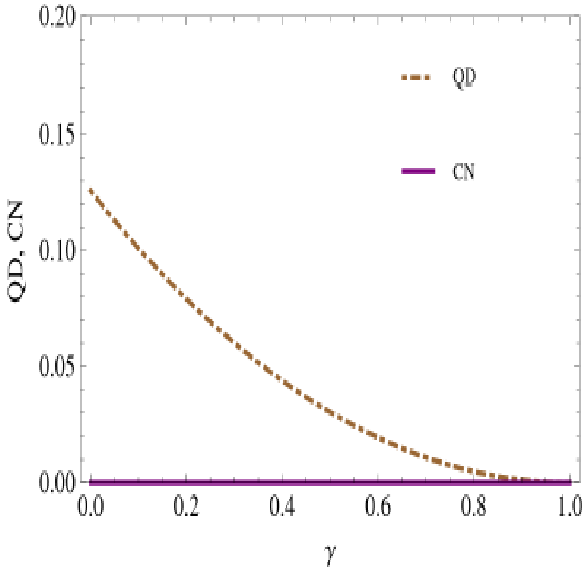
$$|1\rangle_S|0\rangle_E \longrightarrow \sqrt{(1-\gamma)}|1\rangle_S|0\rangle_E+\sqrt{\gamma}\,|1\rangle_S|1\rangle_E$$

$\longrightarrow$

$$E_0=\begin{pmatrix}1 & 0\\ 0 & \sqrt{1-\gamma}\end{pmatrix},\quad E_1=\begin{pmatrix}0 & 0\\ 0 & \sqrt{\gamma}\end{pmatrix}$$

with $\gamma=1-e^{-\Gamma t}$

Using Werner state $\rho(0) = (1-\alpha)I/4 + \alpha|\psi^-\rangle\langle\psi^-|$ (where $\alpha \in [0, 1]$ and $|\psi^-\rangle = 1/\sqrt{2}\,(|01\rangle - |10\rangle)$, spin singlet state) as the initial state, one gets $\rho(t)$

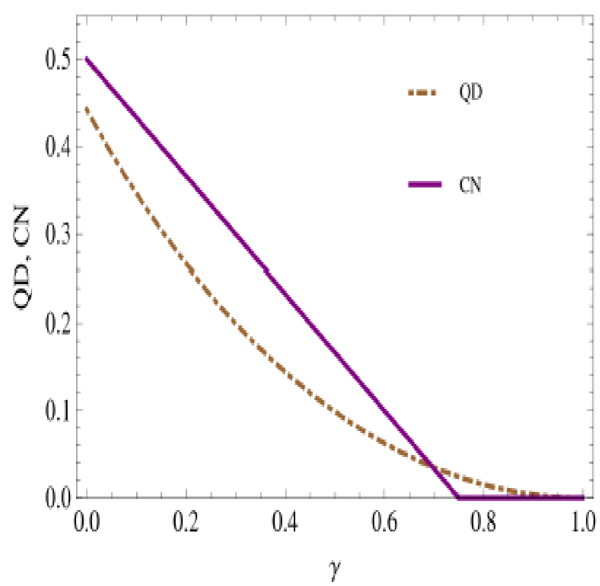


$$CN(\rho_{AB}) = \alpha(3/2 - \gamma) - ½$$

$$Q(\rho_{AB}) = ¼ \{F(a+b)+F(a-b)\} - F(a)/2$$

where $F(x) = x \log_2 x$

$a = (1+\alpha)$, $b = 2\alpha(1-\gamma)$



- The QD vanishes asymptotically with time in both the cases (in agreement with Ref. [6]), but not the CN. CN goes to zero at a finite time for the tetramer and is zero at all *t* for the trimer.

The variation of the quantum discord (QD) and the concurrence (CN) with γ for the AFM ground state of the trimer (top, α = 1/3) and the tetramer (bottom, α = 2/3).

Conclusions

[1] We find that the QD has a non-zero value for both T = 0 and T ≠ 0 in the case of the spin trimer.
[2] The QD jumps in magnitude at the first-order QPT point.
[3] The QD goes to zero only asymptotically with T in both the cases of the trimer and the tetramer which indicates that thermal fluctuations cannot kill the quantum correlations though the latter are reduced in amplitude with higher temperatures.
[4] The two-qubit reduced density matrices at T = 0 for the trimer and the tetramer have the form of Werner states. The QD vanishes asymptotically with time when an initial Werner state is subjected to a dephasing channel.

Sudden Transition Between Classical and Quantum Decoherence (Mazzola et al., Phys. Rev. Lett. 104 200401 (2010)

Experimental detection with an all-optical setup (Xu et al., Nat. Commun. 1 7 (2010))

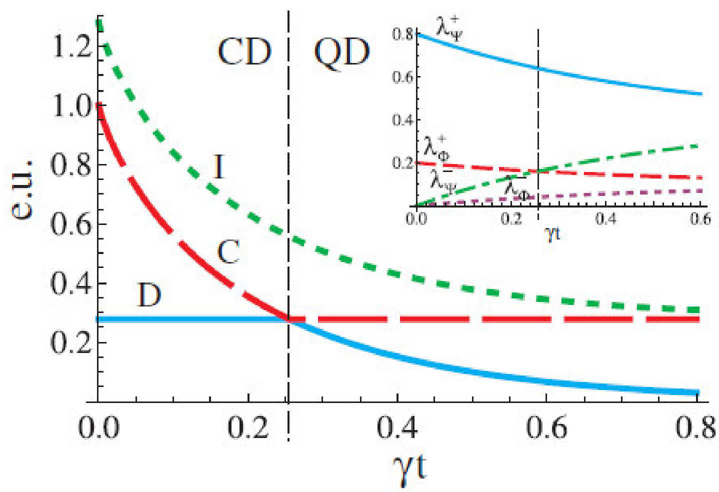


FIG. 1 (color online). Dynamics of mutual information (green dotted line), classical correlations (red dashed line), and quantum discord (blue solid line) as a function of γt for $c_1(0) = 1$, $c_2(0) = -c_3$, and $c_3 = 0.6$. CD and QD stand for the classical decoherence and quantum decoherence regimes, respectively. In the inset we plot the eigenvalues λ_Ψ^+ (blue solid line), λ_Ψ^- (green dash-dotted line), λ_Φ^+ (red dashed line), and λ_Φ^- (violet dotted line) as a function of γt for the same parameters.

First evidence of a quantum property completely unaffected by presence of the environment

Evidence for frozen discord in molecular magnets?

References

[1] M. A. Nielsen and I. L. Chuang, *Quantum Computation and Quantum Information* (Cambridge University Press, Cambridge, England, 2000).
[2] H. Olivier and W. H. Zurek, Phys. Rev. Lett. **88**, 017901 (2001).
[3] L. Henderson and V. Vedral, J. Phys. A **34**, 6899 (2001).
[4] M. S. Sarandy, Phys. Rev. A **80**, 022108 (2009).
[5] S. Luo, Phys. Rev. A **77**, 042303 (2008).
[6] T. Werlang, S. Souza, F. F. Fanchini, and C. J. Villas Boas, Phys. Rev. A **80**, 024103 (2009).

Amit Kumar Pal and Indrani Bose, J. Phys. B: Atomic and Molecular Physics (2011)