Comment on "Quantum Control and Entanglement in a Chemical Compass"

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In the Letter "Quantum Control and Entanglement in a Chemical Compass" [1], Cai *et. al.* study the time evolution of the electron spin entanglement in radical-ion-pair reactions. As one of their main results, the authors calculate the entanglement lifetime, T_E, as a function of the applied magnetic field, reproduced in Fig.1 for convenience.

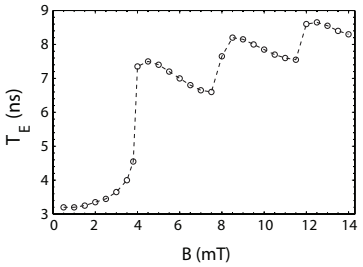

FIG. 1: Figure 2b of [1]

We argue that this result is unphysical, because it leads to a magnetic field estimation much more precise than allowed by fundamental measurement precision limits. The reason is the following. From Fig.1 it is seen that the entanglement lifetime increases discontinuously at $B = 4$ mT. This steep change of T_E with B leads to a very precise estimation of B. Indeed, for a finite signal-to-noise ratio at time $t = 0$, $(S/N)_0$, the precision δT_E of a measurement of T_E is limited [2] by the reaction time T_r, i.e. $\delta T_E = T_r/(S/N)_0$. This is so because molecules, and hence the measurable signal, exponentially disappear with time constant T_r. Thus one cannot measure T_E with any better precision by waiting more than T_r, because there will be no molecules left to do the measurement. Hence the magnetic sensitivity δB, i.e. the smallest measurable change of the magnetic field, is

$$\delta B = \delta T_E/[\Delta T_E/\Delta B] = \frac{T_r/(S/N)_0}{\Delta T_E/\Delta B} \qquad (1)$$

where $\Delta T_E/\Delta B$ is the slope of T_E versus B at a particular value of B. From Fig.1 it is seen that around $B = 4$ mT we have $\Delta T_E/\Delta B \approx 4$ ns/mT. The recombination rate used by the authors is $k = 5.8 \times 10^8$ s^{-1}, leading to a reaction time $T_r = 1/k \approx 1.7$ ns. If we take $S/N = 10$ (the particular value is immaterial), we find $\delta B = 0.04$ mT. Not only is this an overestimate of the magnetic sensitivity δB, but here we have a magnetic field measurement, the precision of which is proportional to T_r, i.e. the shorter the measurement time, the more precise the measurement. This is impossible. This unphysical results comes about because, according to the authors, the slope $\Delta T_E/\Delta_B$ is independent of T_r. This is not the case, as will be now explained.

In reality δB is inversely proportional to T_r. Indeed, a magnetic field measurement is equivalent to an energy measurement, the precision of which is $\delta E = \gamma \delta B$, where $\gamma = 2\pi \times 2.8$ MHz/G. For a measurement time T_r the precision δE is $1/T_r$ [3] improved by the measurement's $(S/N)_0$ ratio, hence

$$\delta B = \frac{1/(S/N)_0}{\gamma T_r} \qquad (2)$$

Thus the magnetic sensitivity actually is about 0.3 mT, i.e. an order of magnitude worse than Cai *et. al.* predict.

The root of the unphysical result presented in [1] is the fact that, according to the authors, the time evolution of the entanglement measure $E(t)$ is induced solely by the magnetic Hamiltonian. The authors have not taken into account intra-molecule spin decoherence [4], which will suppress $E(t)$ [5] and hence T_E will come out to be drastically different. In other words, not taking into account decoherence overestimates the measurement precision, which is a rather established fact in the field of precision measurements.

By including decoherence, the correct scaling of δB with T_r comes about as follows. The decoherence rate is [4] the recombination rate k, and the entanglement decays at least as fast [5], hence $T_E \sim 1/k$. Furthermore, for small magnetic fields the singlet state S is mixed with all triplet states (T_0, $T_\pm$), reducing the entanglement (only S and T_0 are entangled states), whereas for high fields the states $T_\pm$ split away, leaving only S and T_0 to dominate the mixing. The splitting relative to the width k of the reacting singlet state is $\gamma B/k$. Hence $T_E \sim \gamma B/k$, and combining these two arguments we get $T_E \sim \gamma B/k^2$. Thus $\Delta T_E/\Delta B = \gamma T_r^2$, and substituting into (1) we retrieve (2).

[1] J. Cai, G. G. Guerreschi and H. J. Briegel, Phys. Rev. Lett. **104**, 220502 (2010).
[2] This holds for $T_r \lesssim T_E$, which is the case at hand.
[3] S. Boixo, S. T. Flammia, C. M. Caves and J. M. Geremia, Phys. Rev. Lett. **98**, 090401 (2007).
[4] I. K. Kominis, Phys. Rev. E. **80**, 056115 (2009).
[5] S. F. Huelga *et. al.*, Phys. Rev. Lett. **79**, 3865 (1997).