Comment on "Macroscopic Quantum Tunneling in Magnetic Proteins"

The purpose of this Comment is to discuss the implications of some elementary quantum mechanics for Awschalom *et al.*'s observation [1] of a resonance at ~ 1 MHz in ferritin, which they interpret as due to macroscopic quantum tunneling (MQT) of antiferromagnetism. I argue below that a consistent application of their picture requires a large increase in the width and a decrease in the height of the resonance with increasing dc field. Although Awschalom *et al.* now report [2] some such observations, the disagreement is enough (particularly in the resonance width) to continue warranting suspicion about their claim of having seen MQT.

In the picture of Ref. [1], each ferritin particle resonates between two degenerate states in which all the individual Fe^{3+} moments in it are reversed, so that the net residual moment $\mathbf{M}$ ($|\mathbf{M}| = M_0 = 217\mu_B$) follows the order parameter l adiabatically. In current usage, in fact, this picture is that of macroscopic quantum coherence (MQC) between two degenerate states, and not MQT, which describes tunneling out of a metastable well. MQC is generally much harder to see than MQT for several reasons [3], the most relevant here being the need to maintain relatively strict degeneracy between the states.

In Awschalom *et al.*'s experiment, the degeneracy is broken by an external field B. They compare the observed B dependence of their resonance frequency with the formula

$$\nu_{\mathrm{res}}(B) = [\nu_{\mathrm{res}}^2(0) + (M_0 B/h)^2]^{1/2}, \qquad (1)$$

which is obtained if one models the system by a two-state Hamiltonian $(M_0 B \sigma_z + \Delta\sigma_x)/2$, where $\Delta = h\nu_{\mathrm{res}}(0)$. Although the measured $\nu(B)$ does increase with increasing B, the disagreement with Eq. (1) is very large, being an order of magnitude at 2.5 mG, the largest field studied.

In fact, this disagreement is not the main reason for skepticism that one is seeing MQC. There are two other issues that must be considered. First, the mixing between two nearly degenerates states decreases as the departure from degeneracy increases. Thus, the height of the resonance should decrease with increasing B. Second, since the moments $\mathbf{M}_i$ of the individual ferritin particles are randomly oriented, one should have a spread of resonance frequencies and power absorptions, and the observed line shape should be an average.

To estimate the resonance width and height that is expected from the MQC picture, let us write the bias energy for the ith particle as $\epsilon_i = 2\mathbf{M}\cdot\mathbf{B}$, and the Hamiltonian as $(\epsilon_i\sigma_z + \Delta\sigma_x)/2$ [4]. The resonance frequency and the relative power absorption are given by $\nu_i = E_i/h$ and $(\Delta/E_i)^2$, where

$$E_i = (\epsilon_i^2 + \Delta^2)^{1/2}. \qquad (2)$$

For $M_0 B \ll \Delta$, one thus expects an additional linewidth of order $M_0 B/h$, and a modest decrease in the height χ_{max}. In the case $M_0 B \gtrsim \Delta$, we can argue that only particles on or near resonance, i.e., those with $|\epsilon_i| \lesssim \Delta$, will absorb power effectively. Thus we expect the resonance line to extend from $\nu_{\mathrm{res}}(0)$ to approximately $2\nu_{\mathrm{res}}(0)$, and the total area under it to vary as the fraction $\Delta/M_0 B$ of resonant particles. This implies that the height $\chi_{\mathrm{max}} \sim h\gamma_0/M_0 B$, where γ_0 is the width when $B=0$.

Using the value $M_0 = 217\mu_B$ from Ref. [1], one has $M_0 B \simeq h\nu_{\mathrm{res}}(0)$ at $B = 2.5$ mG. At this field, one thus expects an additional width of ~ 500 kHz, and a height, relative to the $B=0$ value, of ~ 0.05. At higher values of B, the linewidth should not change much, but the height should continue to decrease. If instead of $M_0 B$, one uses the observed $\nu_{\mathrm{res}}(B)$ to obtain an effective B-dependent bias, the effects on the line shape are expected to be even more severe.

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[1] D. D. Awschalom *et al.*, Phys. Rev. Lett. **68**, 3092 (1992).

[2] D. D. Awschalom *et al.*, following Comment, Phys. Rev. Lett. **70**, 2199 (1993).

[3] See, e.g., A. J. Leggett, in *Chance and Matter*, Proceedings of the Les Houches Summer School, Session 46, edited by J. Stora, J. Vannimenus, and R. Stora (North-Holland, Amsterdam, 1987).

[4] Strictly speaking, one should also allow Δ to depend on the bias, but this variation is negligible since $M_0 B \ll h\nu_{\mathrm{AF}}$, where ν_{AF} is the antiferromagnetic resonance frequency. (This is $\omega_0/2\pi$ in Ref. [1].) Typically, $\nu_{\mathrm{AF}} \sim 1-10$ GHz.