Comment on "Have Resonance Experiments Seen Macroscopic Quantum Coherence in Magnetic Particles? The Case from Power Absorption"

In a recent Letter [1], Garg makes a case for the origin of the resonance observed in our ac susceptibility measurements [2] on ferritin particles being microscopic (i.e., the tunneling of individual spins) rather than macroscopic quantum coherence (MQC), i.e., the coherent tunneling of all 4500 spins in a particle. In this Comment, we argue that although, as previously noted in Refs. [2] and [3], the MQC interpretation has some troubling features, it provides a far more reasonable explanation of all the data than does the microscopic hypothesis of Ref. [1].

The net magnetic moment M_0, of a ferritin particle, as deduced from dc susceptibility measurements [2], is about $217\mu_B$. Garg [1] argues that in the MQC picture the size of the observed ac resonance requires a moment 7 to 12 times larger. This implies a discrepancy of roughly 50–150 in the power absorption (which varies as M_0^2) for the particles. Though measurements of the absolute, as opposed to relative, magnitudes of the ac susceptibility in the experiments on such tiny particles are difficult and subject to considerable uncertainty, his view is that this discrepancy is large enough to rule out MQC as the source of the resonance. Before accepting this conclusion, however, one must compare the MQC interpretation with the three distinct alternative microscopic explanations he advances:

(i) The tunneling spins belong to the Fe^{3+} ions bound to diamagnetic apoferritin particles, which were mixed [2] with ferritin in the experiments to produce solutions of different concentration. In this scenario, pure apoferritin would display the resonance. However, as a control for the data reported in Ref. [2], we performed susceptibility measurements on pure apoferritin, finding no hint of a resonance.

(ii) The nuclear spins of protons in the ferritin cores do the tunneling. Aside from the daunting problems [1] of explaining the observed size of the resonance in this picture, the small nuclear magnetic moment implies that the resonant frequency should not shift appreciably with magnetic field until fields of 100 G are applied; in fact shifts occur for mG fields, leaving a discrepancy of 10^5.

(iii) Spins of the Fe^{3+} ions in the ferritin cores are the tunneling entities. This mechanism only succeeds [1] in reducing the discrepancy in the power absorption by a factor of roughly 3. To achieve this modest improvement, one is forced to postulate that the Néel temperature for ferritin is 10^4 times smaller than typically quoted values, and that the anisotropy strengths conventionally assumed are too small by 10^2 to 10^3. One is, moreover, again at a loss to explain the sensitivity to magnetic field of the measured resonance frequency: The discrepancy is a factor of 10^2.

Reference [1] also points out that the narrowness of the observed resonance requires a spread of only about 1% in the number of spins in the various ferritin particles. While such strict uniformity is somewhat surprising, there is a basis for rationalizing it: The experiments were performed on particles that had been sifted magnetically in an attempt to narrow the distribution of magnetic moments. The width of the resonance of the sifted particles was significantly less than that of the unsifted ones. It is very hard to understand why such sifting would reduce the resonant width if, as Garg suggests, the resonance reflects the tunneling of individual spins.

In sum, therefore, we find that Ref. [1]'s suggestion of microscopic tunneling presents far more serious difficulties than the (admittedly imperfect) MQC hypothesis.

Finally, Garg's remarks [1] on the resonance observed in our original ac susceptibility measurements [4] on $Fe(CO)_5$ may produce the false impression that we also claimed the origin of this resonance to be MQC. As Ref. [4] and our introduction in Ref. [2] make clear, this is simply not the case. Our data analysis [4] identified fundamental problems with the MQC explanation, notably an observed magnetic field dependence 9 orders of magnitude less sensitive than theory predicts. This gross discrepancy, which Ref. [1] neglects to mention, made clear that the explanation of the data (which remains elusive) did not lie with MQC. We have no quarrel with Garg's calculation on the subject, which confirms this conclusion, only with the implication that we failed to recognize the monumental problems with the MQC picture for $Fe(CO)_5$. It is worth adding that the discrepancy of a factor of 10^{18} he quotes has precisely the same source as the factor 10^9 we pointed out, viz., the extreme smallness, relative to all other energies in the problem, of the observed resonant splitting.

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Received 28 October 1993
PACS numbers: 75.60.Jp, 03.65.Bz, 76.20.+q, 76.30.Fc

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