Comment on "Diameter dependence of ferromagnetic spin moment in Au nanocrystals"

Lin He
Department of Physics, Beijing Normal University, Beijing 100875, People's Republic of China
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Recently, Hori *et al.*[1] reported diameter dependence of ferromagnetism in Au nanocrystals. Their experimental results are systematic and attractive. The authors have shown that the magnetization per nanoparticle surface area decreases with increasing diameter and vanishes at a diameter about 4 nm, as shown in Fig. 6 of Ref. 1. However, in the following, I present arguments showing that their result can be, instead, an apparent behavior resulting from the false analysis of the size dependence of saturation magnetization reported in Fig. 4 of Ref. 1.

In Fig. 4 of Ref. 1, the saturation magnetization of Au nanoparticles was plotted in unit of emu/g. To obtain the diameter dependence of magnetization per nanoparticle surface area, as shown in Fig. 6, Hori *et al.*[1] used the saturation magnetization in Fig. 4 to divide the average surface area of a particle, πD^2 with D the average diameter of the particles. Unfortunately, the magnetization in Fig. 4 is the saturation magnetization of the system. It does not directly reflect the magnetic moments per particle. For a system with constant mass, m, the number of particles, N, of the system is given by $N=m/m_p=m/(\rho\pi D^3/6)$ with m_p the average mass of a particle, and ρ the density of the corresponding material. The total surface area, S, of the system is given by $S=N\times\pi D^2=6m/(\rho D)$. Therefore, the data reported in Fig. 6 of Ref. 1 do not real reveal the magnetization per nanoparticle surface area.

To elucidate this question, I calculate the magnetization per nanoparticle surface area of Au nanocrystals by using the data reported in Fig. 4 of Ref. 1 to divide the surface area S of 1 g Au nanoparticles with different diameter. The result is plotted in Fig. 1. The magnetization per surface area increases linearly with the increasing diameter when the diameter of nanoparticles is smaller than 3.3 nm, rather than decreases linearly with the diameter, as reported in Fig. 6 of Ref. 1. For nanoparticles with diameter larger than 4 nm, the diameter-dependent behaviors differ much from that of the samples with diameter smaller than 3.3 nm, as shown in Fig. 1. The variation of the diameter dependent behavior about 4 nm in Au nanoparticles may be attributed to the spin correlation length of Au.[1]

Figure 2 shows the magnetic moments per particle (left axis) and magnetization per particle (right axis) as a function of the diameter, which are calculated by using the data reported in Fig. 4 of Ref. 1 to divide the number of particles N of 1 g Au nanoparticles with different diameter. Interestingly, the magnetic moments (magnetization) per particle increase monotonic with the increasing diameter. However, for a system with constant mass, m, the number of particles, $N=m/(\rho D^3/6)$, of the system decreases dramatically with increasing diameter. Therefore, the intrinsic diamagnetic signal of Au will cover up the magnetic moments of Au nanocrystals with large diameter.

In summary, we demonstrated that the magnetization per surface area of Au nanoparticles increases with the increasing diameter when the diameter of nanoparticles is smaller than 3.3 nm, rather than decreases linearly with the diameter reported in Ref. 1.

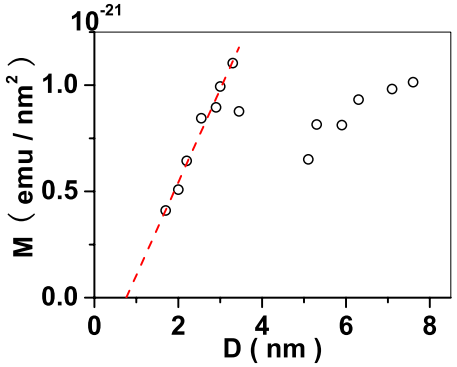

FIG. 1. (Color online) Magnetization per nanoparticle surface area calculated according to the data reported in Fig. 4 of Ref. 1. The dashed line is the guide for the eye.

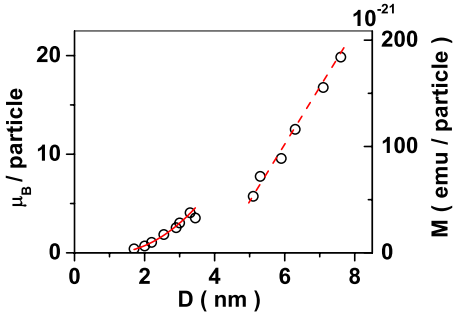

FIG. 2. (Color online) Magnetic moments per particle (left axis) and magnetization per particle (right axis) as a function of the diameter calculated according to the data reported in Fig. 4 of Ref. 1.

[1] H. Hori, Y. Yamamoto, T. Iwamoto, T. Miura, T. Teranishi, and M. Miyake, Phys. Rev. B **69**, 174411 (2004).

