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High-spin tunneling in Fe_8 molecular magnets

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PACS. `75.40.Gb` – Dynamic properties (dynamic susceptibility, spin waves, spin diffusion, dynamic scaling, etc.).
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Abstract. – Results of an experiment on a Fe_8 powder done in the solid state physics laboratory in Orsay have been presented in the literature. In the present contribution, a more simple interpretation of these results is given. Moreover, the Fe_8 powder is studied with a SQUID magnetometer and is shown to be non-oriented.

Results of an experiment on a Fe_8 powder have been presented in the literature [1]. This experiment has been done in the solid state physics laboratory in Orsay and I want to notice two important points:

– The first one is the way the experiment is presented in ref. [1] as the observation of mesoscopic quantum coherence, when it is more simply an EPR experiment that allows to observe the transitions inside the fundamental doublet of the Fe_8 cluster splitted by the magnetic field perpendicular to the easy magnetic axis of this one, as described by Korenblit and Shender [2]. However, it has been shown that the tunneling rate is too small with only an easy axis [3]. A more satisfactory tunneling rate is obtained by considering a second anisotropy constant (eq. (1)). This model describes yOz easy-plane anisotropy with an easy axis along the z-direction in the plane. It has already been considered and the tunneling rate has been calculated without magnetic field [4]. The magnetic susceptibility at high frequency is measured with a resonator in a static magnetic field as in an EPR experiment. The imaginary part of the magnetic susceptibility exhibits two peaks (fig. 1) which are well predicted by the Hamiltonian used to explain the EPR experiments [5, 6]:

$$\mathcal{H} = -DS_z^2 + ES_x^2 - g\mu_{\mathrm{B}}\boldsymbol{B}\cdot\boldsymbol{S}\,, \tag{1}$$

where z and x denote the easy and hard axis, respectively, $S = 10$ and B is the magnetic field. The dotted lines in fig. 1 indicate the peak locations obtained from the numerical diagonalization of the Hamiltonian (eq. (1)) with $D = 0.31\,\mathrm{K}$ and $E = 0.092\,\mathrm{K}$, and the magnetic field perpendicular to the easy axis and parallel and perpendicular to the hard axis, respectively [1].

– The second point is the assertion that "the orientation of the powder was done by solidifying an epoxy (Araldit) with Fe_8 micrometric crystallites buried inside, in a 5.5 T

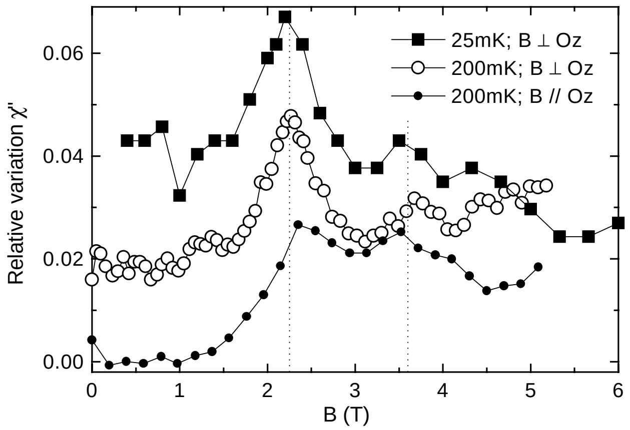

Fig. 1 – From ref. [1]: Imaginary part of the magnetic susceptibility of Fe_8 at 680 MHz as a function of tranverse magnetic field. The dotted lines indicate the peak locations obtained from numerical diagonalization of the Hamiltonian (eq. (1)) with the magnetic field perpendicular to the easy axis using the parameters $D = 0.31\,\mathrm{K}$ and $E = 0.092\,\mathrm{K}$. Since the absolute value of the power absorption is unknown, the curves are arbitrarily shifted in the vertical direction, for clarity.

field at 290 K during 12 hours". This assertion is suspicious because the interaction energy between magnetic ions inside the Fe_8 clusters is much smaller than the thermal energy at room temperature. Therefore, I have measured the magnetization of *the sample that was used in the experiment of ref.* [1] with a SQUID magnetometer at 2 K. The results are reported in fig. 2. It can be seen that there is no significant difference between the magnetization curve for the magnetic field parallel to the powder orientation axis and the one for the magnetic field perpendicular to the powder orientation axis. It is the same in fig. 1, where the main peaks for the two orientation axes appear different on the high-field side but they are similar on the low-field side. So, the difference between the two curves is rather due to a change in the base-line.

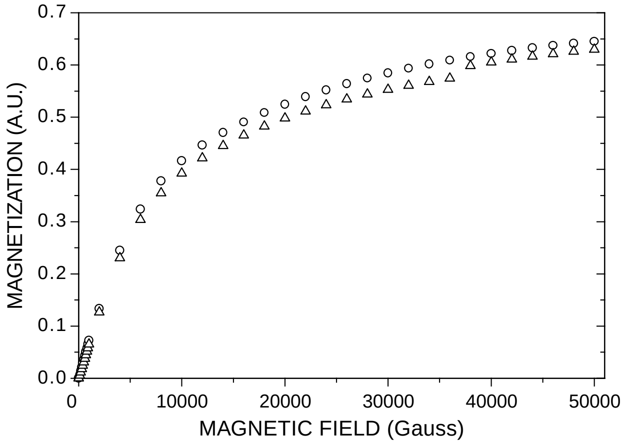

Fig. 2 – Magnetization of the Fe_8 powder at 2 K for the magnetic field parallel to the orientation axis (the empty triangles) and perpendicular to the orientation axis (the empty circles).

To conclude, the susceptibility peaks reported in ref. [1] are observed in a non-oriented powder. They can be explained with tunneling transitions inside the fundamental doublet of the Fe_8 clusters. Only the crystallites that have their easy axes almost perpendicular to the magnetic field take part in the susceptibility peak. This explains why the signal corresponds to a quite small part of the magnetic moments in the sample.

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