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High-resolution detection of resonant frequencies of microwave resonators via magnetic measurements

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An experiment with high spin molecular magnets inside microwave resonators has produced the remarkable observation of sharp peaks in the equilibrium magnetization and the ac susceptibility for the resonant frequencies of these devices. These peaks appear when the magnets are pumped or warmed by microwaves to states of different magnetic moment, and allow one to carry out spectroscopic studies of either the magnetic materials or the resonant device. © *2003 American Institute of Physics.* [DOI: 10.1063/1.1586786]

The basic ideas behind determination of resonant frequencies of a resonator rest on measurement of both the absorbed and reflected energy as a function of the frequency, and the amplitude and phase of the reflection coefficient.[1–3] This implies the use of sophisticated equipment, like network and swept amplitude analyzers, which only work for limited ranges of frequencies. High-resolution measurements of the quality factor Q and frequency are also very important for studying the material properties using resonant techniques.[4,5] This is the case, for example, of the measurement of the surface resistance and reactance of conductors and superconductors or the electric and magnetic susceptibility of dielectrics. Measurement of the microwave power can be realized by using, for example, adiabatic calorimeters, basic bridge circuits and bolometers. We describe an experimental method for quick, high-resolution determination of the resonant frequencies of any resonator based upon the variation of the equilibrium magnetic moment of a magnetic sample placed inside.

In the molecular cluster Mn_{12} acetate ($\mathrm{Mn}_{12}\mathrm{Ac}$) the manganese atoms are tightly coupled to give a constant spin of $S=10$ and below 10 K show magnetic bistability due to the 65 K anisotropy energy barrier between the spin up and spin down states.[6] The occurrence of this strong uniaxial anisotropy yields doubly degenerate ground states in zero field and a set of excited levels in the microwave–infrared range, as has been derived from electron paramagnetic resonance (EPR) resonant experiments.[7,8] The transition probability, Γ, between the spin states at the two wells of the anisotropy barrier U obeys the law $\Gamma=\nu\exp[-U/T]$, where ν is an attempt frequency of the order of $10^6\ \mathrm{s}^{-1}$ and T is the temperature of the experiment. We used in our experiments 2-mm-long 0.5-mm-wide $\mathrm{Mn}_{12}\mathrm{Ac}$ single crystals.[9]

We have investigated several resonant devices, like coils and cavities. In this letter we show data for two resonant loops: the first one is made up of five coils of 0.1-mm-thick 99.9%-pure copper (Cu) wire with a 5.42 mm diameter, and the second one made up of five coils of 0.015-mm-thick superconducting niobium–titanium (NbTi) alloy. We fixed the magnetic samples in the center of the resonant loops and coupled the power to the resonator through a coaxial cable that brought the microwaves from the generator to the sample. We generated electromagnetic radiation using HP83621B synthesized sweepers whose performance was accomplished when they were part of a HP8510C network analyzer system. In the experiments shown here, we swept the radiation from 0.1 to 25 GHz in steps of 0.02 GHz. The radiation power ranged from 0.1 to 300 mW. We placed both the coaxial cable and the resonator inside a commercial sample magnetometer that allows one to change the temperature between 1.8 and 300 K and apply magnetic fields up to $\mu_0H=5$ T.

We characterized the samples magnetically outside and inside the resonator. Both data sets are identical after subtracting the latter the signal due to the resonator. Above the blocking temperature, $T_B\approx 3$ K, the system behaves as a superparamagnet and the equilibrium dc susceptibility follows Curie–Weiss law, $\chi_0=M/\mu_0H=(g\mu_BS)^2/T$, see Fig. 1. Below T_B, the sample exhibits magnetic hysteresis. The inset of Fig. 1 presents the temperature dependence of both the in-phase, χ', and the out-of-phase, χ'', components of the ac susceptibility,[6] with $\omega=57$ Hz the ac frequency. The block-

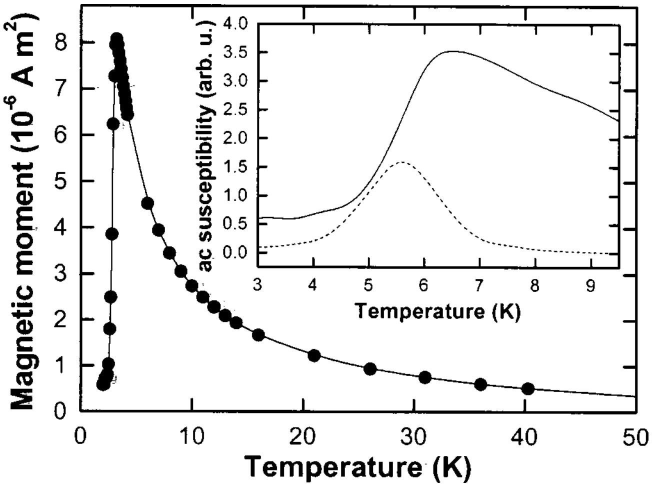

FIG. 1. Temperature dependence of the equilibrium magnetic moment of the $\mathrm{Mn}_{12}\mathrm{Ac}$ sample when we applied magnetic field of $\mu_0H=0.05$ T. The inset shows the thermal variation of the in-phase (solid line) and out-of-phase (dashed line) components of the ac susceptibility ($\omega=57$ Hz).

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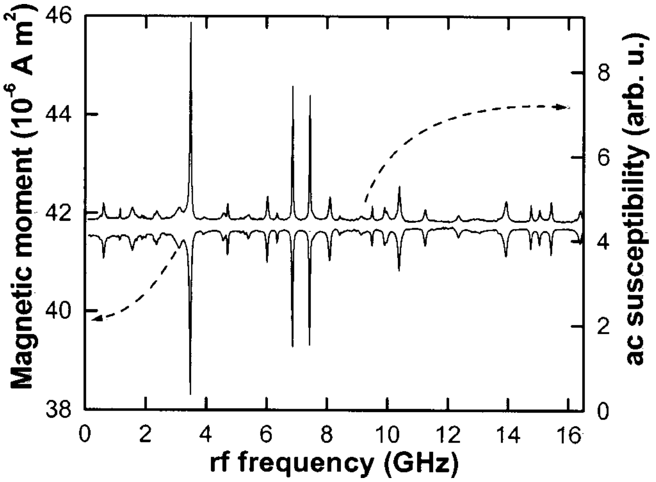

FIG. 2. Equilibrium magnetic moment (left) and real component of the ac susceptibility (right) of the Mn_{12}Ac sample inside the Cu resonant loop at $T=5$ K as a function of the frequency of 300 mW microwave radiation. The magnetic moment was recorded with magnetic field $\mu_0H=0.3$ T, while the ac susceptibility was acquired for ac frequency $\omega=57$ Hz in zero dc field.

ing temperature at this frequency is approximately $T_{B,\mathrm{ac}} \approx 6$ K.

Figure 2 and 3 present the equilibrium magnetic moment of Mn_{12}Ac in the superparamagnetic regime ($T=5$ K) as a function of the frequency of the 300 mW electromagnetic radiation sent, respectively, to the Cu resonant loop and the NbTi resonant loop. The magnetic moment shows sharp decreasing peaks for certain frequency values while there are other frequencies for which it does not change at all (the error in background level is less than 0.01%). In all cases, when we changed the power of the radiation, the intensity of the peaks varied but their positions remained unaltered. Figure 2 also shows the microwave frequency dependence of the real component of the ac susceptibility ($\omega=57$ Hz) of Mn_{12}Ac inside the Cu resonant loop at 5 K and zero dc field. The positions of the increasing peaks of the ac susceptibility coincide completely with those found for the magnetic moment at thermodynamic equilibrium, and the relative variation at the largest peak is 10 times larger than the maximum relative variation of magnetic moment. We also verified that the peaks detected for the NbTi resonant loop coincide with those recorded for the reflection coefficient measured with the HP8510C network analyzer.

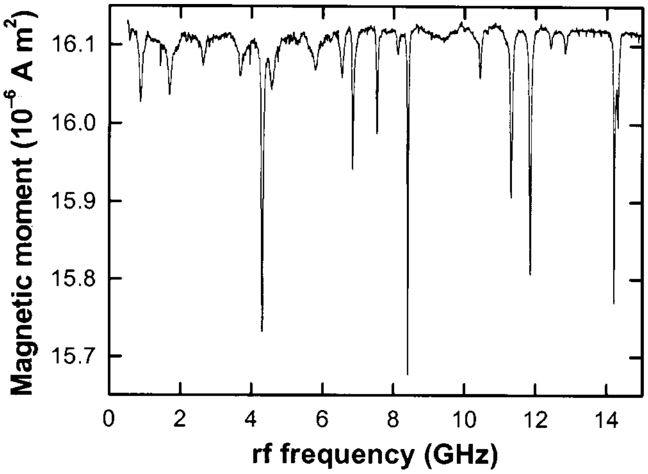

FIG. 3. Equilibrium magnetic moment at $T=5$ K and $\mu_0H=0.1$ T of the Mn_{12}Ac sample inside the NbTi resonant loop as a function of the frequency of 300 mW microwave radiation.

We interpret these results by considering that, when the frequency of the radiation sent to the resonator matches that of a resonant mode, standing waves form and live a certain time, which depends on the Q value of the resonator. Since we are sending radiation during the magnetic measuring time these standing waves continuously appear and disappear, and energy is therefore released. One part of this energy is directly absorbed by the material and the other, which is due to dissipation in the surface of the resonator, will warm the sample. These two effects manifest themselves as variations in the magnetic moment of the sample. When the frequency of the radiation does not match a resonant mode, the radiation is mostly reflected and the magnetic moment of the sample remains constant.

In order to see whether there is microwave photon absorption by the magnetic material inside the resonators, we estimated the ac magnetic field associated with the resonant modes and compared it to the usual ac fields in EPR experiments, which rely on the detection of photon absorption. This field may be calculated as $H_{\mathrm{ac}}^2 \approx 8\pi E/V$, where V is the volume of the resonator and E is the energy stored within. This energy is $E=QP/2\pi f$, where Q is the quality factor of the resonator, P is the time-averaged power loss and f is the microwave frequency.[1] Using typical values for these magnitudes, we obtained $H_{\mathrm{ac}} \approx 0.01-0.1$ Oe, a value that compares fairly well to that of the usual ac fields in EPR experiments.[10] We should therefore expect to have quantum transitions between levels whose spacing matches the energy of the photons. The energy absorbed by molecular (mostly vibrational) levels should flow towards phonon degrees of freedom and affect the population of spin levels via heating. In the case of spin levels, the photons absorbed by levels corresponding to different precessional motion of the total spin of the molecular magnet could increase the population of excited levels and reduce the total magnetic moment of the sample.

In conclusion, we have shown and discussed the use of magnets to determine the microwave frequencies of a resonator. The results shown here, together with those obtained using other loops and cavities as well as different magnetic materials (paramagnets and gadolinium) and magnetic sensors (Hall probes), suggest that our method is fast and very precise in determining the microwave resonant frequencies, the Q values and the radiation power.

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[1] M. Sucher and J. Fox, *Handbook of Microwave Measurements* Vols. I, II, and III (Polytechnic Press of the Polytechnic Institute of Brooklyn, New York, 1963).

[2] O. Klein, S. Donovan, M. Dressel, K. Holezer, and G. Grüner, Int. J. Infrared Millim. Waves **14**, 2423 (1993); S. Donovan, O. Klein, M. Dressel, K. Holezer, and G. Grüner, *ibid.* **14**, 2459 (1993); M. Dressel, O. Klein, S. Donovan, and G. Grüner, *ibid.* **14**, 2489 (1993).

[3] S. Hill, P. S. Sandhu, C. Buhler, S. Uji, J. S. Brooks, L. Seger, M. Boon-

man, A. Wittlin, J. A. A. J. Perenboom, P. Goy, R. Kato, H. Sawa, and S. Aonuma, Proc. SPIE **2842**, 296 (1996).
[4] J. Mollá, A. Ibarra, J. Margineda, J. M. Zamarro, and A. Hernández, IEEE Trans. Instrum. Meas. **42**, 817 (1993).
[5] D. Kajfez and A. Gundavajhala, Electron. Lett. **29**, 1936 (1993).
[6] E. M. Chudnovsky and J. Tejada, *Macroscopic Quantum Tunneling of the Magnetic Moment* (Cambridge University Press, Cambridge, UK, 1998).
[7] S. Hill, J. A. A. J. Perenboom, N. S. Dalal, and T. Hathaway, Phys. Rev. Lett. **80**, 2453 (1998).
[8] A. L. Barra, D. Gatteschi, and R. Sessoli, Phys. Rev. B **56**, 8192 (1997).
[9] T. Lis, Acta Crystallogr., Sect. B: Struct. Crystallogr. Cryst. Chem. **B36**, 2042 (1980).
[10] A. Abragam and B. Bleaney, *Electron Paramagnetic Resonance of Transition Ions* (Clarendon, Oxford, UK, 1970).