Magnetic resonance force microscopy: Nonlinear processes and influence of relaxation times

L. Lenci,* D. Bertolini, D. Friselli, M. Martinelli, and G. Scalari
Istituto per i Processi Chimico-Fisici del Consiglio Nazionale delle Ricerche, Via Moruzzi 1, I-56124 Pisa, Italy
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The phenomenon of the micromechanically detected magnetic resonance is interpreted in terms of nonlinear processes at magnetic resonance. A close analysis in the frequency domain of the irradiation and detection scheme shows that the technique corresponds to a multiple irradiation with one or more couple of frequencies separated by ω_c and to the detection of the longitudinal component of magnetization oscillating at ω_c. The study of longitudinal detection of magnetic resonance allows the direct measurement of the spin-lattice relaxation time of samples. Samples of Mn^{2+}:MgO prepared in order to obtain a mixture of spin systems with very different relaxation processes were studied by electron-spin-resonance experiments with micromechanical detection: measurements evidence a very strong rejection of the system with lower longitudinal relaxation time. Direct confirmation of the theoretical interpretation is obtained; in addition the microscopy technique increases its ''contrast'' capability, adding the possibility of determining maps of samples based on the distribution of both concentration and longitudinal relaxation times of spin systems.

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I. INTRODUCTION

Since the discovery of the magnetic-resonance phenomenon, nearly all the experiments used as a physical observable, for the detection of resonance signals, the electromagnetic field interacting with the sample in magnetic-resonance condition.[1,2] Only isolated experiments accounted for the effects of magnetic resonance on some parameters of the sample itself, such as the temperature.[3] A complete study of the effects connected at magnetic resonance to absorption by the sample of the angular momentum rather than of the energy of photons was on the other hand developed in 1960s;[4,5] this allowed the observation of a macroscopic torque determining the oscillation of a paramagnetic torsional pendulum. The peculiar effects of the low intensity static magnetic fields used and of many photon transitions were evaluated and interpreted.[6]

A renewed impulse to novel methods for the detection of magnetic resonance came from the proposal, the detection, and the development of the micromechanical observation of magnetic resonance.[7–15] This methodology presents several interesting features, combining aspects of atomic force microscopy with magnetic-resonance processes in presence of field gradients. The main objectives of this approach are the increased sensitivity and spatial resolution directed to the study of both single nuclear- and electronic-spin systems; a different scanning probe microscopy arises named magnetic-resonance force microscopy (MRFM). Technological efforts were addressed chiefly to enhance the sensitivity of the detector cantilever and the value of field gradient applied to the samples. Accordingly, a force noise floor of the order of $10\hat{}\{-18\}$ N was demonstrated with special cantilevers at low temperatures,[16–18] and field gradients as high as 2.5×10^5 T/m were employed.[19]

More recently increased attention was paid to the possibility of studying the relaxation processes in both electron and nuclear systems.[20,21] In particular, it was demonstrated that in particularly strong gradient and in high saturation conditions the Bloch equations no longer account for the resonance phenomenon; on the other hand special combinations of pulse and continuous-wave techniques allowed the study of transverse and longitudinal relaxation times in nuclear MRFM of micron-size crystals.[21] The possible effects due to the presence of systems with different relaxation times were also discussed.

The analysis of the MRFM technique shows however that the force signal is connected to the dynamics of the longitudinal component of magnetization. A simple study of the experimental procedures shows in addition that the different modulations impressed in the radiation and magnetic field where samples are embedded, required to stimulate the mechanical resonance of the cantilever, correspond in the frequency domain to multiple irradiation. The MRFM experiment as a whole resembles the nonlinear magnetic-resonance technique named longitudinally detected electron-spin resonance (LODESR).[22–24] This technique, thoroughly studied several years ago, implies a double (or multiple) transverse irradiation of a sample at magnetic resonance; in this nonlinear condition the longitudinal component of the sample magnetization shows oscillations at the difference in frequency between the irradiating waves and harmonics.[22,24] The analysis of the line shape shows that the signal at the first harmonics has the same width as for the normal electron-spin-resonance (ESR) signal but its intensity is proportional to the saturation factor, and then supplies direct information about the spin-lattice relaxation time also in low saturation condition.[24] This simple and informative technique was then extensively used for studying spin-lattice relaxation processes in a number of physically different samples.[25,26]

This paper reports about the study of MRFM experiments in the light of LODESR phenomenon. Impressive results show the capability of MRFM in the separation of contributions to the signal from spin systems with different longitudinal relaxation times. This suggests the possibility of obtaining in a simple way a relaxation microscopy.

Section II shortly recalls the fundamentals of MRFM experiment with some comments about the detection methods.

The representation of the irradiation scheme in the frequency domain is discussed, too.

Section III, starting from the above irradiation scheme, reports an overview of the theory of LODESR phenomenon. The line shape is calculated, for simplicity, for the case of a spin $S=1/2$, useful for the demonstration of the phenomenon; the more complicated case $S=5/2$ relevant to the experiments is not reported here since a quantitative comparison with the line shape is not the subject of this paper. The physical equivalence with the MRFM process is evidenced.

Section IV describes the essentials of the apparatus used for ESR and micromechanical detected ESR experiments, the preparation of the sample, and spectra obtained with different experimental configurations. The evidence of tight connection with LODESR experiments is discussed.

Section V finally presents some conclusions about the achievements of the work and indicates some useful perspectives for the implementation of a relaxation-resolved MRFM spectroscopy.

II. THE MICROMECHANICAL DETECTION OF MAGNETIC RESONANCE

The method of mechanical detection of magnetic resonance exploits the observation of mechanical effects produced in a spin system at resonance. Such effects can be observed either in the sample itself or in a mechanical device integral with the sample. The first method was investigated in a series of works in 1960s where the sample operated as a torsional pendulum acted on by the angular momentum of photons absorbed at resonance.[4–6] This method is effective mainly at low values of the static magnetic field. The second method, following the suggestion of Sidles in the early 1970s (Ref. 7), implies the interaction of the sample with a micromechanical lever used for detecting the mechanical effects exerted on the sample at magnetic resonance. These effects can be either the torque corresponding to the angular momentum of photons[11] or the pressure due to the magnetization of the sample when inserted in a field gradient.[8,9,14] Most of the experiments deal with the last technique: the sample is supported on a cantilever, submitted simultaneously to a magnetic field and to a field gradient in the same direction *z*. This gives a force F_z exerted on the cantilever due to the resonant volume V of the sample given by

$$F_z = VM_z \frac{\partial B_z}{\partial z}. \tag{1}$$

The magnetization M_z of the sample can be changed by magnetic-resonance processes produced by submitting the sample to resonant irradiation; the torsion of the cantilever changes accordingly. The mechanical resonance of the lever can be usefully employed to amplify the phenomenon, provided that the magnetic-resonance processes are modulated at a frequency coincident with the resonance frequency of the lever itself. The related oscillations of the cantilever are detected with optical methods (interferometric or optical lever methods). In some cases the inverted configuration,[27] where the cantilever supports a small magnetic particle generating the field gradient and scanning across the sample placed in fixed position, is profitably used.

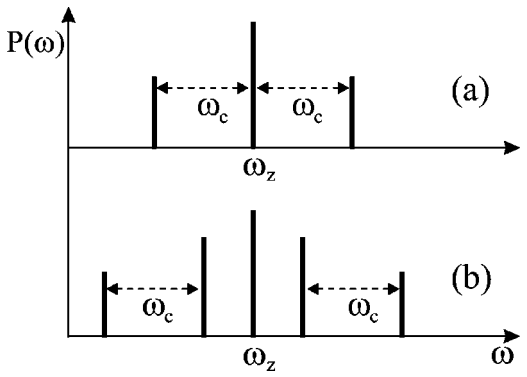

FIG. 1. Effective radiation spectrum seen by the sample when (a) only the amplitude of the electromagnetic wave is modulated; (b) the amplitudes of the static and radio-frequency fields are modulated.

The main features of this technique are the higher sensitivity, compared to classic inductive methods, and the possibility of investigating the spatial distribution of spins inside the sample, due to the peculiar presence of the field gradient.

MRFM could achieve, in perspective, the requested sensitivity to detect the signal from a single electronic or nuclear spin. This feature could be useful to the implementation of a solid-state quantum computer.[28–30]

In general the modulation process is drawn by magnetic-field modulation, often combined with modulation or periodical switching of the irradiation amplitude. If indeed the field is modulated at the frequency ω_c, characteristic of the cantilever, a spurious signal is usually impressed to the detection device. This drawback can be avoided by performing a double modulation (field-amplitude or field-field) at frequencies ω_{m1} and ω_{m2} such as $\omega_{m1}-\omega_{m2}=\omega_c$ or $\omega_{m1}+\omega_{m2}=\omega_c$. Since the sample at resonance has a nonlinear behavior, it acts like a frequency mixer giving all the intermodulation products of the incoming radiations; obviously the cantilever among all the available mechanical solicitations selects the one at frequency ω_c. The overall irradiation scheme is usefully considered in the frequency domain; the stick spectrum of Fig. 1 represents the effective radiation spectrum seen by the sample for both the amplitude modulation alone and the double modulation cases.

The condition represented in Fig. 1 corresponds to a simple radiation amplitude sinusoidal modulation at frequency ω_c [Fig. 1(a)] and a double modulation field-amplitude at two frequencies that differ from each other by ω_c [Fig. 1(b)]; ω_z is the Larmor frequency of the spin system. If the wave amplitude is controlled by a switch, the structure of the spectrum includes a larger number of components but the frequency difference between each couple of spectral components remains the same.

It must be remarked that the actual irradiation scheme and the use of the observable M_z in MRFM experiments indicates a close connection with the phenomenon of LODESR; in the following section the theoretical predictions of LODESR method will be shortly summarized and used to

calculate the MRFM signal. Successively a comparison of experiments with the two procedures will evidence the conceptual identity of the two techniques; special attention will be paid to the effects of relaxation times that disclose different perspectives to the MRFM procedure.

III. THE LONGITUDINAL DETECTED MAGNETIC RESONANCE

Longitudinal detection of electron spin resonance (LODESR) is a technique proposed and observed several years ago;[22–24] the theoretical interpretation of the phenomenon was progressively refined while experiments showed the information potential of this technique.[25,26] The basic phenomenon investigated involves a spin system interacting with more than just one nearly resonant wave at magnetic resonance. Some of the drawbacks present in the usual Bloch equation formalism in the rotating system were avoided by using the second-quantization formalism for an arbitrary number of waves interacting with the magnetic system. By this way the different Hamiltonians (of the system, of the radiations, and of the system-radiation interactions) become time independent; the interaction operates on eigenstates of the system-radiation and only eigenstates nearly degenerate must be considered.[31] Resonances are obtained by a diagonalization of the whole matrix including the effects of the interactions on the states direct product of radiation and magnetic system states.[31] The line shape of the different resonances were obtained by using the density-matrix formalism and the phenomenological description of the relaxation processes by the transversal and longitudinal relaxation times of Bloch equations. A long series of papers deeply elucidated the different phenomena observed mainly in electron paramagnetic-resonance conditions.[22–24] Here only some generalities of the theory will be recalled in order to understand the peculiarities of the phenomenon and the connections with MRFM and to predict some very important properties of the micromechanically detected magnetic resonance.

The simplest LODESR phenomenon involves a spin system (in the case considered here an electron spin S_z, z being the direction of the static magnetic field) irradiated with two monochromatic transverse electromagnetic waves oscillating at frequencies ω_r and ω_s, both nearly resonant with the Larmor frequency ω_z of the system.

In the ''dressed atom'' formalism the total Hamiltonian H is written as

$$H = H_S + H_R + H_I , \tag{2}$$

where the Hamiltonian H_S of the isolated spin system, the Hamiltonian H_R of the free field, and the interaction Hamiltonian H_I, in $\hbar = 1$ units, are given, respectively, by[31]

$$\begin{aligned} H_S &= \omega_z S_z , \\ H_R &= H_R^{(r)} + H_R^{(s)} = \omega_r a_r^\dagger a_r + \omega_s a_s^\dagger a_s , \\ H_I &= H_I^{(r)} + H_I^{(s)} = g\mu_B (\omega_r/2V)^{1/2} (a_r^\dagger + a_r) S_x \\ &\quad + g\mu_B (\omega_s/2V)^{1/2} (a_s^\dagger + a_s) S_x , \end{aligned} \tag{3}$$

$a_r^\dagger$, $a_r^\dagger$ and a_r, a_s being the creation and annihilation operators of the two waves, V the volume where the electromagnetic (e.m.) waves are present, and μ_B the Bohr magneton. In the following, the case $S_z = 1/2$ will be considered for simplicity. In order to study the resonances, a nearly degenerate manifold of the Hamiltonian $H_S + H_R$ must be considered, represented on the basis of the eigenstates $|m, n_r, n_s\rangle$, where m is the spin z component occupation number and n_r, n_s the photon occupation number for the two electromagnetic waves. The set of degenerate states is then

$$\begin{gathered} |-1/2, n_r + 1, n_s\rangle \\ |1/2, n_r, n_s\rangle \\ |-1/2, n_r, n_s + 1\rangle \; |+1/2, n_r - 1, n_s + 1\rangle \\ : \qquad ; \end{gathered} \tag{4}$$

the states for simplicity can be labeled with a single index, starting from the states $|0\rangle, |1\rangle$ corresponding to the central states of the succession defined in relation (4). The representative matrix of the total Hamiltonian [see Eq. (2)], apart from a constant diagonal term $(n_r\omega_r + n_s\omega_s - \omega_z)/2$, is written as

$$\begin{pmatrix} .. & \cdots & & & & & & & \\ .. & \Delta_1 - 2\Delta_2 & \lambda_r & & & & & & \\ & \lambda_r & -\Delta_2 & \lambda_s & & & & & \\ & & \lambda_s & \Delta_1 - \Delta_2 & \lambda_r & & & & \\ & & & \lambda_r & 0 & \lambda_s & & & \\ & & & & \lambda_s & \Delta_1 & \lambda_r & & \\ & & & & & \lambda_r & \Delta_2 & \lambda_s & \\ & & & & & & \lambda_s & \Delta_1 + \Delta_2 & .. \\ & & & & & & & \cdots & .. \end{pmatrix} , \tag{5}$$

where $\Delta_1=\omega_s-\omega_z\approx\omega_r-\omega_z$, $\Delta_2=\omega_r-\omega_s$, and λ_r and λ_s are the matrix elements of H_I between the couples of states of the degenerate set for the *r* and *s* waves, respectively. The resonances of the system are calculated by diagonalizing the matrix (5) and considering the degenerate eigenvalues.

The line shape of each resonance is obtained by using the density-matrix formalism. The density operator for the total interacting spin-radiation system can be conveniently written as $\rho=\rho_0+D$, where ρ_0 is the density operator at the thermodynamic equilibrium and D takes into account the relaxation mechanisms. The operator D obeys the evolution equation[31–33]

$$\frac{i}{\tau}D=[H,D]-\frac{\beta[H_R,H_I]}{\mathrm{Tr}\{\exp[-\beta(H_S+H_I)]\}}, \tag{6}$$

where i/τ is the relaxation superoperator and $\beta=1/k_BT$ the Boltzmann factor. Equation (6), represented on the basis (4), is an equation system for the different matrix elements $D_{i,j}$. Because of the high number of present photons, states corresponding to the same eigenvalue of the spin momentum are all physically equivalent, so the recurrence rule $D_{i,j}=D_{i+k,j+k}$ holds, where $k=2(S_z+1)$. The i/τ operator can be phenomenologically represented by

$$\left[\frac{i}{\tau}D\right]_{i,j}=\begin{cases}\dfrac{i}{T_1}D_{i,j}, & i=j\\ \dfrac{i}{T_2}D_{i,j}, & i\neq j.\end{cases} \tag{7}$$

Consequently, after the trace calculation the system (6) becomes[26,27]

$$\frac{i}{T_{i,j}}D_{i,j}=[H,D]_{i,j}-\frac{\beta}{2S_z+1}[H_R,H_I]_{i,j}. \tag{8}$$

It must be noticed that the matrix element $D_{i,j}$ corresponds to a process involving $|i-j|$ photons. In the search of elements accounting for the first harmonic of M_z (oscillating at the frequency $\Delta_2=|\omega_r-\omega_s|$), the solution of the system (8) must be obtained for elements of the type $D_{i,i+2}$. Exploiting the recurrence rule it is sufficient to calculate the term D_{02}. For this only the terms corresponding to a process involving a number of photons compatible with experiments can be retained in relation (8) and analytical approximate solutions are obtained.[22–24] By assuming $\lambda_r=\lambda_s$ the response of the system for the component of M_z oscillating at the frequency Δ_2 is given by[25]

$$M_z(\Delta_2)\propto\frac{H_1^2T_1T_2}{\frac{i}{T_1}+\Delta_2}\left(\frac{\omega_r}{\frac{i}{T_2}+(\omega_0-\omega_r)}+\frac{\omega_s}{\frac{i}{T_2}+(\omega_s-\omega_0)}\right). \tag{9}$$

Equation (9) presents some interesting features. In fact the information content depends on the procedure of the experiment. If the value of ω_0 is changed (i.e., the magnetic field is swept) and Δ_2 is kept fixed two resonances are observed, whose amplitude depends on the product T_1T_2 while the linewidth depends only on T_2. If on the contrary ω_0 is kept fixed and Δ_2 is varied, the spectrum observed depends on the values of the relaxation times; in particular when the condition $T_1\gg T_2$ holds (condition fulfilled in the majority of solid-state samples) the amplitude of the detected line still depends on the product T_1T_2 and the line shape is connected only to T_1, since variations of the signal driven by T_2 occur in a far larger frequency range. It is worth noting that both field and frequency swept experiments give information about the longitudinal relaxation time. The LODESR signal even connected to a nonlinear process was observed also for rather low values of the saturation factor.[22,24]

It must be pointed out that the theoretical findings of the preceding section were referred to the case of a spin-1/2 system. If $S\neq1/2$ (as for Mn^{2+} ion that has $I=5/2$ and $S=5/2$), the matrix elements of the field-spin interaction Hamiltonian H_I between a state with spin quantum number M_S and a state with spin quantum number $H_I\pm1$ are obtained from the corresponding matrix elements for $S=1/2$, just by multiplying them with the factor $[S(S+1)-M(M\pm1)]^{1/2}$. However the complete calculation of the line shape for the more complicated case $I=5/2$ and $S=5/2$ is over the findings of this work.

LODESR experiments were carried out with both a cw two-wave scheme and a single wave sinusoidally modulated.[34] LODESR spectroscopy has been performed also in a pulsed way, using only one source of the e.m. wave. Describing the e.m. pulses with a Fourier series and looking into the magnetic field $B(t)$ of the e.m. wave of frequency ω_0 amplitude modulated with a period T_m leads to

$$B(t)=\tilde{B}\sum_{n=-k}^{k}b_n\sin(\omega_0+n\omega_m)t, \tag{10}$$

where $\tilde{B}$ is the amplitude of electromagnetic wave and b_n are the coefficients giving the amplitude of different spectral components. The irradiation with pulses spectrally corresponds to an irradiation with a set of harmonics spaced by a frequency equal to the rate of repetition of the pulses. Figure 2 shows the stick spectrum of irradiation schemes in LODESR experiments with different configurations. A direct inspection of Figs. 1 and 2 suggests the basic identity of the irradiation scheme for the MRFM and LODESR cases, provided that $|\omega_r-\omega_s|=\omega_c$.

In addition, M_z being in both experiments (MRFM and LODESR) the physical observable of interest, it is straightforward to conclude that the above spectroscopies give the same information about the properties of the magnetic system.

IV. THE LODESR BY MICROMECHANICAL DETECTION

The LODESR detection technique is performed inductively (a coil pair with the axis parallel to the static magnetic field detects the signal relative to the M_z oscillation) while the signal related to the mechanical detection of the magnetic resonance observed in an MRFM experiment is just the longitudinal component of the magnetization transduced by the

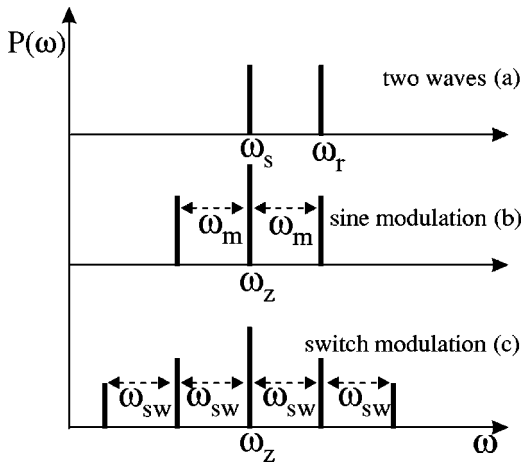

FIG. 2. Effective radiation spectrum seen by the sample when (a) two waves oscillating at frequency ω_s and ω_r are applied, (b) the amplitude of one wave is modulated by a sine wave, and (c) the amplitude of the one wave is modulated by a switch (on-off) control.

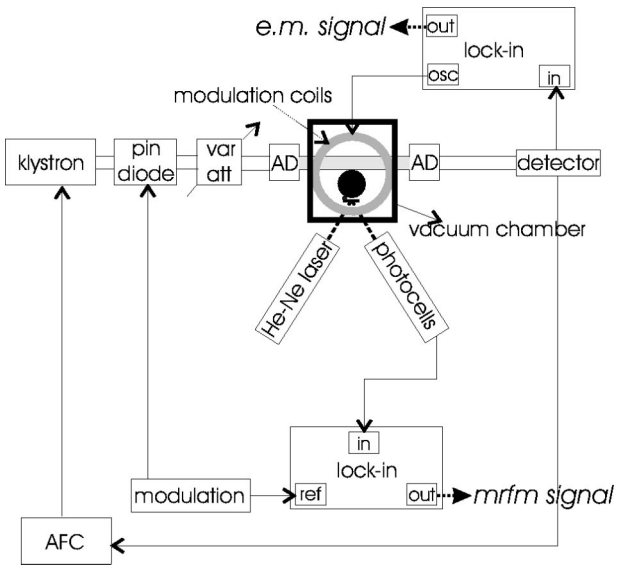

FIG. 3. Block diagram of the overall experimental apparatus.

mechanical oscillator used for the force detection instead of the coil.

To investigate experimentally the similarities of LODESR and MRFM techniques, a series of measurements were performed employing the 23 GHz apparatus described in detail in Ref. 14. The spectrometer is a multipurpose, home-built instrument capable of detecting the electron paramagnetic-resonance phenomenon in three ways: usual electromagnetic detection, force detection (MRFM sample-on-cantilever operating mode), and torque detection. The latter technique takes inspiration from a series of works of the 1960s (Ref. 4) and is described also in a recent paper concerning measures at low fields (400 MHz).[11]

The present spectrometer is based on the use of a dielectric resonator working in a whispering gallery resonance mode (WGM) configuration[35,36] that allows a simple optical access to the region where the cantilever is placed and that shows very good characteristics at the frequency of interest and, in perspective, also at higher fields and frequencies. The cantilever motion is detected by an apparatus based on the optical lever method, implemented to work with distances between the laser source and the cantilever of the order of 30 cm; a recent upgrade of the experimental setup allows the system to operate under vacuum (at a pressure of about 10 mTorr) increasing the sensitivity of the instrument by a factor $(Q_{\mathrm{vac}}/Q_{\mathrm{air}})^{1/2}$, Q being the mechanical merit factor of the loaded cantilever. The magnetic-field gradient is generated by a small iron cylinder 1 mm in diameter and 4 mm long, placed in the proximity of the cantilever by a three-axis micropositioning translator.

Experiments were performed modulating on-off the intensity of the e.m. field at the frequency of mechanical resonance of the cantilever using a *p-i-n* diode switch. The block diagram of the apparatus is reported in Fig. 3.

Measurements were performed, using the three different detection schemes, on a paramagnetic sample constituted by a micrometer-size $(60\times60\times100\ \mu\mathrm{m})^3$ grain of MgO doped with $MnCl_2$. The specimen was prepared by impregnation of magnesium oxide powder with manganese chloride, whose weight was 7% in respect to the MgO one. The obtained compound was dried at 110 °C, mixed, and heated at 600 °C in air for 1 h and then fired at 1000 °C for 5 h, following the procedure reported in Ref. 23. The result of this procedure was to carry out a sample in which two distinct phases were present: some of the Mn^{2+} ions diffuse through the microcrystals of MgO and give a paramagnetic spectrum consisting of six sharp hyperfine lines ($S_z=5/2$, $I_z=5/2$).

The major part of the Mn^{2+} ions remains in a precipitate phase and originates a paramagnetic spectrum consisting of one line strongly broadened by dipolar interaction. Time and temperatures of the sample handling affect the overall observed line shape. In particular it must be pointed out that the relaxation times of the two species contributing to the ESR signal are by far different, the concentration of the two phases and then the relaxation interactions being very different in the two cases.

The sample prepared with the above procedure was glued on the tip of a commercial silicon nitride cantilever (from Park Scientific Instruments) and then placed in the spectrometer in close proximity of the dielectric resonator operating in whispering gallery modes. By this way the resonating radiation wraps the sample.

Figure 4 shows the ESR signal electromagnetically detected; the signal as usual is in derivative shape, being obtained by modulation of the static field by a pair of coils in Helmholtz configuration and phase detection of the signal coming from the detector. The large, broad line is due to the precipitated phase of Mn^{2+} ions. The slight distortion of the large broad line is due to some paramagnetic impurities present in the resonator (a cylinder of alumina), having the *g*-factor value very close to the one of Mn^{2+}. Arrows indicate the position of five of the six lines related to the hyperfine interaction of the Mn^{2+} ions diffused in microcrystals; the other line occurring in the most intense part of the broad spectrum is not easily observable.

The observation of the micromechanically detected ESR

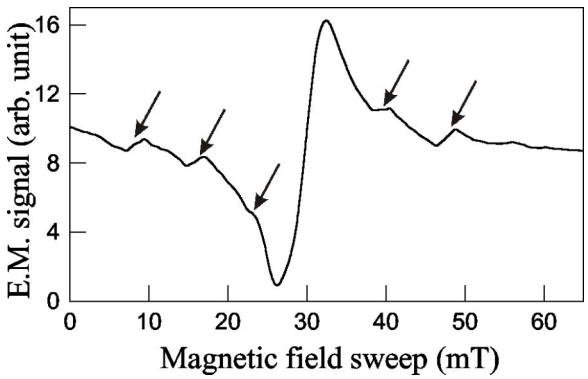

FIG. 4. ESR signal of a grain of MgO doped with $MnCl_2$ electromagnetically detected.

signal of the same sample is obtained simply by pulling down the pressure at the sample up to 10 mTorr, switching the microwave power on-off at a frequency of 705.35 Hz (the resonance frequency of the micromechanical oscillator integral with the sample) and activating the optical oscillation detection.

Figure 5 shows a spectrum micromechanically detected from the same sample as in Fig. 4. The shape of the line is nonderivative because the parameter modulated is the microwave field amplitude.[2] The spectrum appears completely different in comparison with the previous one: in fact the six hyperfine lines are well evident while the large broad line completely vanished. The result is identical to the one shown for a similar sample in Ref. 23. In that case the comparison was between an ESR spectrum and a LODESR spectrum performed on the same sample. This is not surprising if one considers that in both cases (LODESR and micromechanical detection) the signal intensity is proportional to the longitudinal relaxation time T_1 as predicted in previous paragraphs.

It must be pointed out that the hyperfine lines in the case of micromechanical detection (see Fig. 5) are broadened because of the presence of the magnetic-field gradient as an essential element of the technique. The spectrum reported in Fig. 6 for comparison is obtained with the same sample, but acquired with an increased value of the field gradient, carried

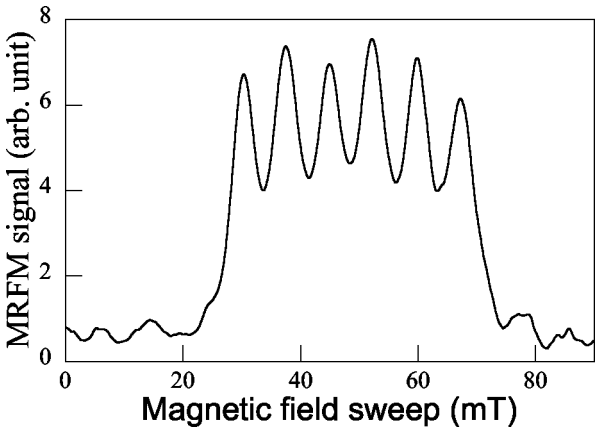

FIG. 5. ESR signal of a grain of MgO doped with $MnCl_2$ force detected.

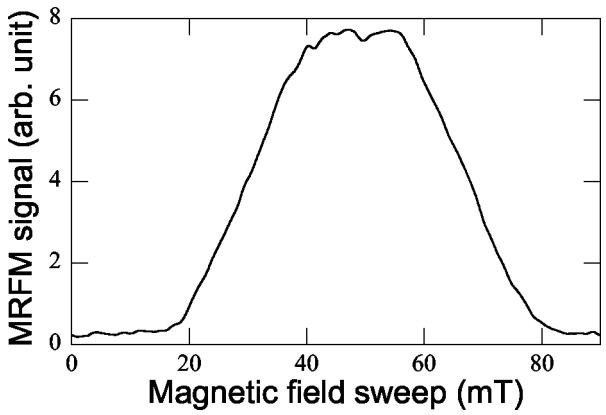

FIG. 6. ESR signal of a grain of MgO doped with $MnCl_2$ force detected with high gradient magnetic-field value.

out by reducing the distance between the iron cylinder and the cantilever integral with the sample: it the broadening of the linewidth is visible, while the central line is still undetectable.

Measurements have been performed with different intensities of the irradiating wave. Figure 7 reports results obtained detecting the first harmonic (the signal oscillating at the mechanical frequency of the cantilever) of the detected signal: the amplitudes of the signal confirms, as expected from theory, the linear dependence of the first harmonic signal intensity on the power of the irradiating wave.

To obtain a comprehensive description of phenomena related to the micromechanical detection of the paramagnetic resonance the torque detection method[4] was also investigated with the same sample. This method is based on angular momentum conservation, and detects the macroscopic torque acting on a magnetized sample during an ESR experiment as a consequence of absorption of angular momentum of photons. In fact the angular momentum of the photons exchanged during the resonance process reveals itself as a macroscopic torque, parallel to the static field $B_{0,}$ acting on the sample and detected in this case by the cantilever. The experimental setup is very similar to the case of the force detection, it is sufficient toremove the field gradient source and

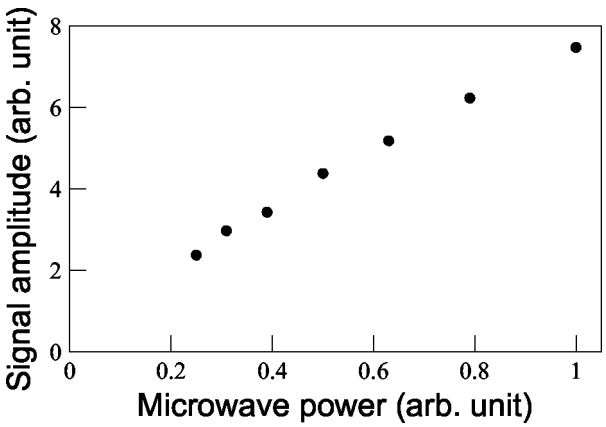

FIG. 7. First-harmonic amplitude of the ESR force detected signal.

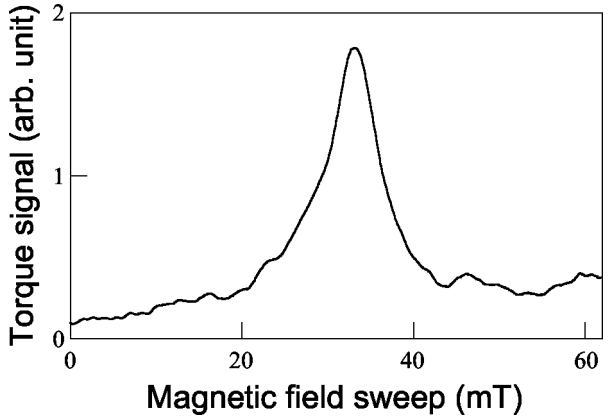

FIG. 8. ESR signal of a grain of MgO doped with $MnCl_2$ torque detected.

to rotate by $\pi/2$ the cantilever in order to detect the cantilever deflection due to the torque. This procedure implies the detection of the transversal component of the sample magnetization[5] whose intensity and shape are connected to the transversal relaxation times T_2. This is evidenced by the spectrum reported in Fig. 8 where the torque ESR signal of the same sample used for previous experiments shows essentially only the single broad line previously observed by the standard electromagnetic detection. The signal relative to the Mn^{2+} diffused in MgO microcrystals in this case is not visible, due to the fact that the torque method is less sensitive at high frequencies, having a signal-to-noise ratio that depends linearly on the field and making this technique particularly appealing at low fields.

In conclusion MRFM technique turns out completely equivalent to the LODESR spectroscopy from a conceptual point of view giving a rich information about the different relaxation processes of studied materials.

V. ACHIEVEMENTS AND PERSPECTIVES

The results here presented give an insight in the interpretation of MRFM phenomenon. The achievements of this work can be summarized as follows.

(1) The process underlying the MRFM spectroscopy has a well distinguished nonlinear character.

(2) The theoretical approach used for the case of LODESR spectroscopy completely accounts for the effects observed with the MRFM method.

(3) In both the above methodologies the role of relaxation times was clarified.

Different applications can be envisaged, and the capabilities of that spectroscopy are enhanced, allowing a more deep study of all the systems where the relaxation processes present a spatial distribution. As examples liquid-crystal systems with partial spatial orientation, solid-state systems with stresses, and systems with concentration gradients can be quoted. In conclusion the considerations reported here account for increased effectiveness of the MRFM technique that shows an improved contrast capability.

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*Author to whom correspondence should be addressed. Email address: lenci@ipcf.cnr.it

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