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NEW SCANNING MILLIMETER WAVEBAND ESR-MICROSCOPE WITH LOCALIZED MAGNETIC FIELD

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Abstract

The design of the new scanning ESR-microscope, which is able to detect magnetic particles smaller than $0.015\times3\times0.025$ mm^3 by the method of Electron Spin Resonance in 30-45 GHz frequency band is presented. The static magnetic field in the working zone of the microscope is created by the use of the special magnetic field concentrator unit. Mechanical shifting of the specimen provides the scanning of the static magnetic field on its surface. The first experimental data, which demonstrate the efficiency of the device operation, are presented and analyzed. The possibility of the ESR-microscope design for the frequency band up to 140 GHz is discussed.

Keywords: millimeter waveband, Electron Spin Resonance (ESR) microscopy, open resonator, magnetic field concentrator.

1. Introduction

An incredible speed with which the results of investigations of magnetic nanostructures [1] are converted to a practice (high density disks, position sensors etc.) stimulates further studies of these structures in extra-high (EHF) [2]- and infrared (IR) [3]- frequency bands. At the same time, a lot of new methods were involved and developed in the course of these researches. The Electron Spin Resonance (ESR) technique is among the most informative methods because of its high sensitivity to the magnetic structure of material under study. The equation of motion for an electron spin in crossed alternating and static magnetic fields ($\vec{h}_{EHF} \perp \vec{H}_{ST}$) leads to the known resonance frequency-resonance field relation [10]:

$$h\nu_{res} = g\beta H_{res} \quad (1)$$

where h is the Plank's constant: ν_{res} is the resonance frequency: H_{res} is the resonance magnitude of the applied external static magnetic field $\vec{H}_{ST}$; β is the Bohr magneton; g is the g-factor or the spectroscopy splitting factor (as a rule for paramagnets $g \approx 2.00 \pm 1\%$). As it follows from (1) the values ν_{res} and H_{res} are connected unambiguously.

The sensitivity of the ESR technique increases drastically with the corresponding increase of the frequency up to dozens and hundreds gigahertz. However the successful application of this method to magnetic nanostructures meets two main obstacles.

1. In the millimeter waveband the ESR method requires strong static magnetic fields, which in turn are connected with cumbersome and expensive magnetic systems. For instant as it follows from (1) in order to detect the ESR signal at $\nu \approx 100$ GHz one should create a static magnetic field with the strength $H \approx 35.6$ kOe.

2. To realize the study of magnetic nanostructures we should obtain ESR conditions in the specimen's area of nano-size dimension, that is smaller than at least 10^{-2} mm^2. This requirement results from the fact that the properties such as Giant Magnetic Resistance/Impedance [1] are usually manifestation of magnetic interactions, which take place between nanoparticles with dimensions 1nm–100 μm.

The methods of nondestructive study of magnetic nanosystems attract much interest today so the elaboration of new special ESR-technique for these purposes is very topical. In our previous paper [4] we have suggested the new ESR resonator cell, which does not require a complicated and bulky magnetic system to study magnetic nanostructures in the millimeter band. This cell can be used effectively for microscopy researches connected with application of ESR technique, or for ESR - microscopy. This paper describes the results of the first microscopy experiments with application of ESR technique and the design of the scanning ESR–microscope operating in the millimeter waveband with spatial resolution better than $0.02 \times 0.1 \times 1$ mm^3.

2. The design of the ESR-microscope and its parameters. Experimental results

The term Electron Spin Resonance Microscope (correspondingly ESR-Microscopy) has been introduced several years ago [5, 6 etc.]. It means the device, which allows detection of ESR response from a very small part (dozens of square micrometers scale) of magnetic (paramagnetic) specimen surface. As a rule the distinctive feature of such device is the frequency band $\nu \approx 10$ GHz and the corresponding value of applied static magnetic field H = 3-4 kOe (to provide the required frequency). To ensure micrometer spatial resolution, a hole is made in the wall of 3 cm (X-band) cavity resonator of spectrometer. The hole diameter is smaller than cut-off diameter. A specimen, under study is introduced into the exponentially damping resonance field, which exists in the vicinity of this hole. So only a small surface area of the specimen is illuminated by this damping electromagnetic field and only this area determines the ESR response detected by the microscope. Then a stepping mechanical scanning provides a mapping of the distribution of paramagnetic centers on the specimen surface.

The described device possesses two intrinsic shortcomings. They are following:

1. A very low operational frequency;
2. Very large overall dimensions, which are governed mainly by the dimensions of electromagnet (1.5-2 m).

In this paper we present a design of the new ESR-microscope, which

1. Operates in the millimeter waveband;
2. Possesses small overall dimensions of about $7 \times 10 \times 20$ cm.

Experimental results obtained with the help of this microscope are also presented below.

The advantages of new microscope as compared with convenient ESR-microscopes were reached because of new basic operational principle. The principle is, that the strong static magnetic field, which is indispensable for operation of the device in the millimeter waveband, concentrated by special magnetic concentrator unit in a small spatial area.

2.1. Magnetic field concentrator unit

Let us consider first the features of the magnetic system, which are essential for the realization of new ESR-microscope. Our design is based on the possibility to create very high strength of static magnetic field H_{ST} in small spatial area. The field is generated only with the help of special configuration of highly anisotropic permanent magnets. This idea was substantiated in our previous papers [4, 7 and 8] and here we shall consider only the details of the particular field concentrator unit. The magnets are fabricated from $SmCo_5$ and possess giant anisotropy field up to 300 kOe. Their magnetization also has rather high magnitude $Ms \approx 750$ G. As the experimental unit has been described in details in [4], here we present only its design (Fig.1a) and the dependence of static magnetic field H_X on X and Y (Fig.1 b,c).

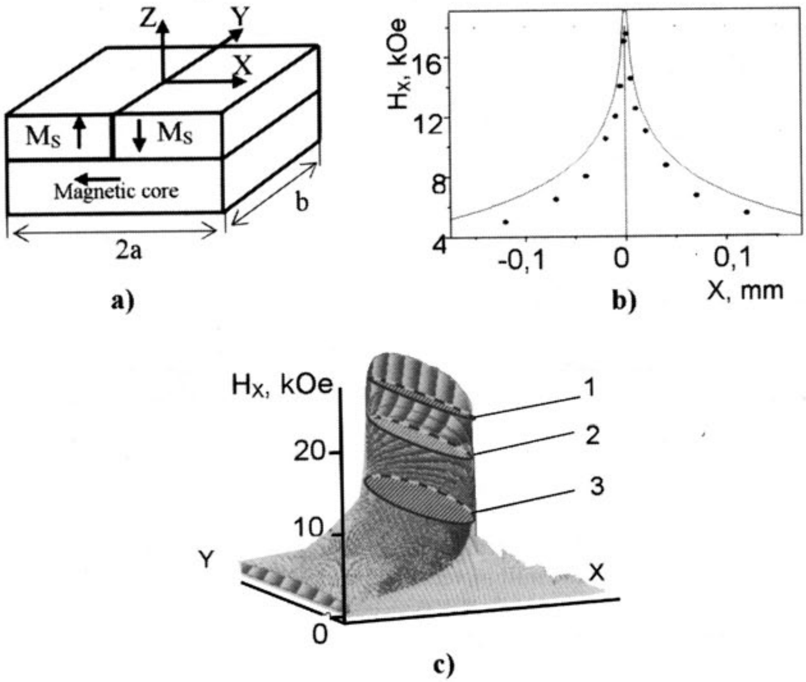


Fig.1. The static magnetic field concentrator unit. a) the flow-chart; b) experimental data (dots) and calculations (solid lines) for H_X component in the center of magnet along OX-axis; c) calculations for $H_X(X,Y)$ on surface of the magnets (Z = 0). The oval-like slices correspond to the ESR-conditions: 1- H_{res}=25 kOe and ν_{res}=56.41 GHz; 2- H_{res}=20 kOe and ν_{res}=45.13 GHz; 3- H_{res}=10 kOe and ν_{res}=22.56 GHz.

As it was stressed in [7, 8] the necessary condition for occurrence of magnetic fields with such high strength was the giant anisotropy of the constituent magnets. In particular the high strength (15-20 kOe) of localized field can be reached in the system of magnets with antiparallel magnetization direction shown in Fig. 1a.

The general formula describing H_X - component in the vicinity of the OY axis is [8]:

$$H_X(X,Z)=M_S\left[\ln\left(a^2-2aX+X^2+Z^2\right)-2\ln\left(X^2+Z^2\right)+\ln\left(a^2+2aX+X^2+Z^2\right)\right] \quad (2)$$

For $r \equiv (X^2+Z^2)^{0.5}/a < 0.01$ this expression has a simple form:

$$H_X \approx 4M_S \ln(X/a) \quad (3)$$

Note that according to more detailed computations the component H_X only weakly depends on Y in the central part of the magnet for the space of several millimeters- Fig. 1c.

The dependence for $H_X(X)$ is presented in Fig. 1b together with experimental dots for M_S=750 G. One can see that the strength of the magnetic field component H_X has a very sharp dependence of the blade-type. The schematic drawing of the resonance field slices $H_{res} = H_X$ for various field values is shown in Fig. 1c. For magnets with dimension $a \approx 20$mm [4] the field $H_X \geq 15$ kOe is confined inside a cylindrical volume with radius $r < 100$ µm, at the same time the higher fields (up to 25 kOe) are confined within noticeably lesser volume ($r \approx 8$ µm).

2.2. The experimental researches

A block-diagram of the millimeter waveband ESR-microscope is presented in Fig.2. It consists of BWO oscillator, operating in 37.5-53 GHz band; P-I-N diode switch; isolator; magic-T; resonance cell; millimeter-wave detector; amplifier & lock-in detector; voltmeter.

A signal generated by BWO oscillator is modulated by P-I-N switch at 1 kHz and irradiates the sample in the resonance cell (Fig.3). operating in a reflection mode configuration. A reflecting signal is detected, amplified and sent to the lock-in detector. The voltage produced by the lock-in detector is registering by A/D voltmeter.

The resonance cell based on Fabry-Perot resonator (Open Resonator/OR) is used in semi-confocal arrangement (Fig.3a) and is tuned to resonate on TEM_{008} mode. The loaded Q-factor equals $Q \approx 1500$. Optimal configuration of open resonator is close to confocal geometry. The spot of the resonance field (the area in XOY plane where the field intensity decreases less than in "e"-times in comparison with maximum) in OR is presented by known Gauss-distribution with its center in the center of plane mirror. The experimentally measured diameter of this spot is approximately 20 mm.

The flat bottom mirror is manufactured from polished $SmCo_5$ magnetic field concentrator described in the previous chapter. The slot between magnets is disposed just in the center of such plane mirror.

The open resonator is excited through the slot-type coupling element, made in the top mirror. This provides the plane polarization of the resonance field in the resonator. The magnetic component (h_{EHF}) of the resonance electromagnetic field is directed along OY-axis. So the ESR condition ($\vec{h}_{EHF} \perp \vec{H}_{ST}$) is satisfied.

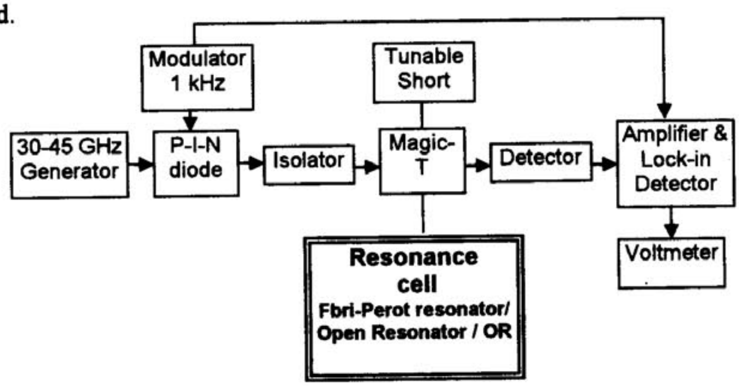


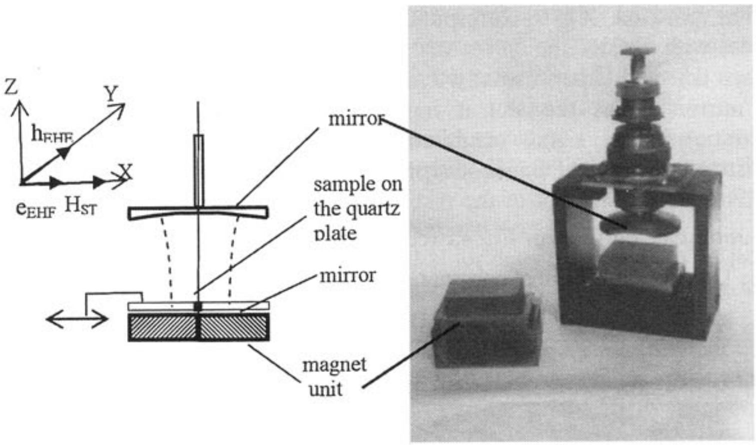
Fig.2. Block-diagram of the ESR-microscope.

The specimen is fabricated as a thin ribbon and sticked to the center of polished quartz plate, which is placed on the bottom mirror in such a way, that the specimen appears both in the vicinity of maximum of the static magnetic field [4] and in the maximum of magnetic component of the resonance electromagnetic field. The quartz plate with the specimen can be scanned mechanically with the help of the mechanical driven positioner.

It was estimated [4] that the static magnetic field at $Z < 15$ μm reaches the magnitude $H_X > 15$ kOe in the operating zone of OR. This magnitude corresponds to ESR conditions for the frequency $\nu > 43$ GHz. Note that these ESR-conditions are created not in the whole specimen but only in a narrow strip-shaped area with dimensions $\delta X \times \delta Y \approx 15$ μm×3 mm.

Special experiments, which fulfill the ESR-signal detecting, have been performed. During such test experiments we used as a small perturbation element two various ribbons of the same size and manufactured from:

1. The nonmagnetic material (aluminum):
2. The reference specimen.



a) **b)**

Fig.3. *The resonance cell of ESR-microscope of the millimeter waveband a) sketch, b) overall view.*

As a reference specimen we used the ferromagnetic ribbon of quick hardened $Fe_{40}Ni_{40}B_{20}$ (metal glass), which was much studied by us earlier [9]. This material is quite convenient for testing goals because it has no planar anisotropy and possesses well- defined magnitude of the saturation magnetization $M_S = 934$ G. The thickness of the ribbon is h = 25 μm. The length of the ribbon (l) equals 5 mm, and its width (d) is the variable parameter of the experiment (d=0.2 mm, 0.4 mm, 1.0 mm). Note that only a part of the specimen finds itself in strong static magnetic field. This part with dimensions $\delta X\times\delta Y\times\delta Z \approx 15\ \mu m\times 3\ mm\times 15\ \mu m$ can be treated as a bar-shaped specimen of finite dimensions.

Therefore the frequency of the electron spin resonance (or, of the ferromagnetic resonance/ FMR in the given case) can be calculated by the known Kittel formula (4) for the specimen of arbitrary shape [10]:

$$\nu_{res} = \frac{g\beta}{h}\left\{\left[\left(N^{(X)} - N^{(Z)}\right)M_S + H_{res}\right]\left[H_{res} + \left(N^{(Y)} - N^{(Z)}\right)M_S\right]\right\}^{\frac{1}{2}} \qquad (4)$$

where M_S is the saturation magnetization of the ferromagnet; $N^{(X)}$, $N^{(Y)}$, $N^{(Z)}$- are demagnetization factors along X, Y, Z – axis correspondingly. Formula represents the generalization of (1) on the case of ferromagnet and allows estimating the static magnetic field value in the site of specimen location.

In Fig.4 the function (U), which characterizes the signal magnitude in the OR is shown versus the position both of the reference ($Fe_{40}Ni_{40}B_{20}$)- sample and of the Al- probe centers relatively to the slot, while their scanning across the slot. The position X = 0 corresponds to the case when specimen's center coincides with the slot.

It was detected experimentally that when Al- specimen is scanned along the plane mirror across the slot it inputs only standard "small perturbation" (dissipation) in the signal magnitude (U). The U(X)-function in this case demonstrates the typical Gauss- distribution about 20 mm (δX=±10 mm) width, which is equal to the spot of the resonance field in OR. The small part of this distribution can be seen in Fig.4a (curve 4) for |dX| ⌋ 1.5 mm. It is naturally that the U-function is almost constant at such small distance.

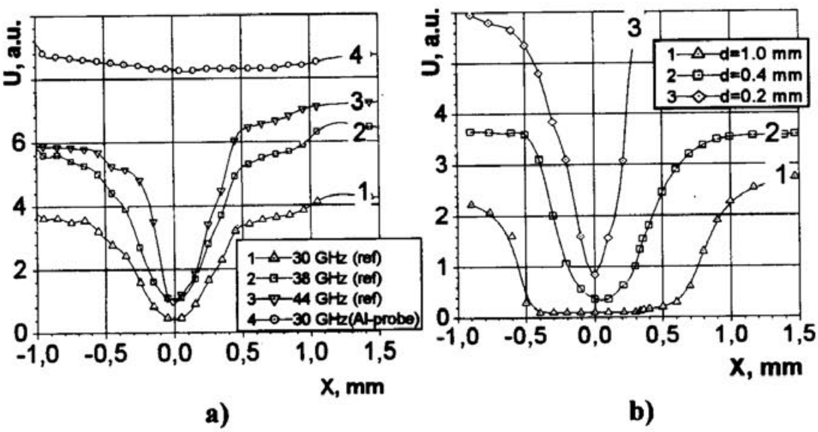


Fig.4. ESR-response from the reference samples. The signal value versus the resonator cell while scanning samples along X-axis:
a) frequency (ν) as the parameter for d=0.4 mm: 1- ref. sample ν=30 GHz, 2- ref. sample ν=38 GHz, 3- ref. sample ν=44 GHz; 4- Al-sample ν=30 GHz;
b) width (d) as the parameter for ν=30 GHz: 1- d=1.0 mm; 2- d=0.4 mm; 3- d=0.2 mm;

Note, that Al specimen should input larger perturbation into the resonance field of OR because the conductivity of Al is approximately 50 times greater than the conductivity of the reference specimen Fe-Ni-B. However for the case when the reference specimen is scanned along the X-axis - a deep well in the signal magnitude distribution U(X) (curves 1-3) can be seen. Its minimum is situated at X = 0 (just in the position where center of the reference sample coincides with the slot).

In Fig.4a the distribution of signal magnitude is shown for the specimen d = 0.4 mm wide at various frequencies. According to the figure the FMR response becomes smaller and the signal becomes weaker as the frequency increases. At the same time the well of the function U(X) becomes narrower. This can be readily explained by the narrowing of the space area (in X-direction) occupied by the static resonance magnetic field H_{res}, as the frequency increases.

To estimate the spatial resolution we detected the ESR response from reference specimens with various widths (Fig.4b). It is easy to see, that the width of the well in X-direction precisely corresponds to specimen's width. This fact proves the efficiency of the ESR microscope.

The calculations based on (4) showed, that for detected frequency of the FMR response, ν_{res} = 44 GHz, the static magnetic field in the vicinity of the slot (in the operating zone of the ESR-microscope) comprised $H_{res} \approx$ 19 250 Oe.

Finally let us estimate the theoretical limit of the operating frequency for the ESR-microscope. Note that values 35 - 36 kOe represent the estimated maximum of static magnetic field, which can be produced by contemporary magnetic concentrator units. So according to (1) the Electron Spin Resonance signal (for paramagnet specimen) can be detected at frequencies up to ν = 100 GHz. For the plane shaped ferromagnetic specimen with the high enough magnetization saturation values ($M_S \approx$ 1.2 - 1.4 kG as a rule) the operating frequency of ESR-microscope can reach, according to (4), magnitudes of order 120-140 GHz.

3. Conclusions

1. The first experiments with new ESR-microscope with localized magnetic field have been performed in the millimeter waveband and main characteristics of the device have been studied.
2. It was proved the possibility of creating the magnetic field with intensity 19 250 Oe and the area of its localization smaller than 0.05 mm^2 by the use of special field concentrator.
3. The two-mirror open resonator has been successfully used as a resonator cell of the microscope. The ESR response from the reference specimen was detected and analyzed in the frequency band ν = 30-44 GHz.

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