About the Possibility of Identification of Hydrocarbon Deposits with the Help of NMR

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Abstract. The main purpose of this article is to review the theoretical prerequisites of nuclear magnetic resonance (NMR) application in tasks of search and exploration of hydrocarbon deposits. The identification peculiarities of hydrocarbon deposits in a weak magnetic field of the Earth were analyzed. The necessity of using highly directional antennas for providing greater sounding depths as well as the use of absorption spectra of the sought-for minerals as sounding signals were identified and justified.A variant of practical implementation of such a system was examined and examples of practical application of the innovative NMR technology were provided.

Keywords: Nuclear magnetic resonance; highly directive/superdirective antennas; Kirlian effect; Larmor frequency

1 Introduction

Nuclear magnetic resonance is a phenomenon of resonant absorption and reradiation of radio frequency electromagnetic energy by substance with nonzero magnetic moments of nuclei in an external constant magnetic field. Nonzero nuclear magnetic moment have nuclei *1H, 2H, 13C, 14N, 15N, 19F, 29Si, 31P* and others.

At the moment it is difficult to specify a field of natural sciences, which would not use NMR to some extent.

NMR methods are widely used in chemistry, molecular physics, biology, medicine and related sciences - that is where the study of substance make up and its molecular structure is required.

In physics NMR has introduced unique information on thermodynamics of spin systems, localization and mobility of nuclei, atomic groups and molecules in crystal lattice. In chemistry NMR has introduced information about the structure and dynamic stereochemistry of many important classes of compounds, exchange processes etc. Effective analytical and structural-chemical methods for medicine, biochemistry, pharmacology, chemical industry were developed on this basis.

NMR has found its place in applied geophysics during creation of magnetometers for magnetic survey (Hill,1960; Nalivayko et al.,1962). Of particular note is the spin-echo method, which allows to determine the process speed of spin-lattice processes and spin-spin interactions. Imaging log (MRIL) was introduced in 1991 by NUMAR (Coates et al.,1991) based on the method of magnetic resonance. Halliburton and Schlumberger have created NMR logging devices, which are successfully used for logging of oil wells.

Radiofrequency impulses are sent to a reservoir in the process of NMR logging and inbetween them the decline of response is measured from those of hydrogen protons, which entered into resonance at a given magnetic field. Here in fact takes place the method of 'direct' survey of reservoir characteristics conducted in the wellbore.

In accordance with (Coates et al.,1992), NMR logging uses the idea of NMR imaging that was originally implemented in medical magnetic resonance imaging (MRI). Only in MRI the object is placed inside the device and in NMR logging it is done on the contrary, the device is placed inside the object - in a wellbore.

A method that also deserves attention is MRS (Magnetic Resonance Sounding) used for search of underground waters at depths of up to 150 m. Measurements are taken remotely from the ground surface and in the Earth's magnetic field – i.e. we see an actually functioning system of NMR for split site instrumental devices and the survey object.

Let us question whether it would be possible to opt for the MRS development pattern and apply it for identification of hydrocarbon deposits. Unfortunately not. There are several major reasons for this:

- hydrocarbon deposits dip much deeper than underground fresh water, thus a simple power increase of transmitter of excitation signals for soil sounding at 5 - 7 km – is a prospectless way.

5   - another issue is that the chemical composition of the oil is incomparably more complex than that of water. According to [Pokonova et al., 1984], oil is a mixture of circa 1,000 individual substances, most of which are liquid hydrocarbons (> 500 substances), sulfur compounds (about 250 substances), nitrogen (> 30 substances) and oxygen compounds (about 85 substances, and organometallic compounds (vanadium, nickel and others). Other components are dissolved hydrocarbon gases, water, mineral salts (mainly chlorides), solutions of salts of organic acids, mechanical admixtures etc. Therefore, the

10   resonance excitation by one sinusoidal frequency is a rather unreliable method.

- the following reason lies in the fact that by exciting all fluid protons, it is difficult to distinguish relaxation of oil and water in the Earth's magnetic.

Moreover, NMR will be carried out in a relatively weak magnetic field of the Earth. The average field density on the surface of the Earth is approximately $5 \cdot 10^{-5}$ T, which is significantly less than the power of magnets for NMR

15   spectroscopy.

Each of these factors is essential and requires its own analysis for the formation of scientific and technical approach to implementation of the direct method of identification and survey of hydrocarbon deposits from the surface of the earth.

Let us consider at first the theoretical background of NMR implementation in conditions listed above.

2 Theoretical Background of Remote Sounding of Interior using NMR

20   Nuclear magnetic resonance is one of the main methods of quantum physics. Primarily magnetic resonance methods include nuclear magnetic resonance, electronic paramagnetic resonance (EPR) and nuclear quadrupole resonance (NQR). These methods provide unique information for physicists and chemists about molecular structure and properties of substances and are powerful diagnostic methods in modern medicine.

NMR discovery became possible thanks to the fact that existence of magnetic moments in many nuclei was

25   experimentally established, which was predicted by Wolfgang Pauli back in 1924 during explanation of hyperfine structure of atomic spectra (Pauli, 1924).

Transitions between the sublevels of Zeeman splitting due to interaction of the summary nuclear magnetic moment with external magnetic field were observed for the first time in 1938 by Isidor Rabi.

In 1946 two groups of American physicists from Stanford and Harvard Universities led by Felix Bloch (Bloch,1946) and

30   Edward Purcell (Parcell,1946) discovered the phenomenon of NMR in condensed matters. Later in 1950 H. Dehmelt and H. Krueger received NQR signals. Shortly double spin-spin, spin-photonic and spin-phononic resonances were developed.

As a result of these outstanding achievements in the field of NMR and EPR obtained by scientists from different countries, there are a number of tried-and-true methods on study of nuclear relaxation, molecular structure etc.

Rapidly developing method of NMR spectroscopy has become multi-nuclei providing the ability of analysis on all

35   magnetic nuclei of the D. Mendeleev's element periodic table. In this case information about the nucleus of this type is extremely accurately associated with intra- and intermolecular structural and dynamic situations.

The important factors for our consideration that shape the unique resonance absorption spectra of substances are the Zeeman effect and the so-called 'chemical' shift.

Zeeman effect (Pieter Zeeman was a Dutch physicist who received the Nobel prize for his discovery in 1902) is the split

40   of lines of atomic spectra in a magnetic field (fig. 1).

The resulting absorption spectrum of a substance is significantly enriched with new components while combining atoms into molecules. The number of lines in the NMR spectrum is determined by the amount of chemically non-equivalent nuclei of this grade and constants of spin-spin interaction of these nuclei.

5 In our case in a weak magnetic field of the Earth we can observe an anomalous Zeeman effect when spectral atomic lines are split into a much larger number of components. As a result spectra of atom absorption and their survey become accordingly more complicated. In some cases the resulting spectra (NMR spectra $_1$H, $_{13}$C, $_{19}$F) may contain hundreds (and even thousands) of resonance lines.

Chemical shift in NMR is an NMR signal bias depending on chemical composition of substance due to shielding of an external magnetic field by electrons of atoms.

10 The phenomenon of chemical shift was originally discovered in 1951 in ethanol spirit *CH₃CH₂OH*. The phenomenon of separate resonance of protons contained in the molecule of the substance was obtained. That is protons have different frequencies of own NMR in various substances. The same atomic nuclei in various environments in molecule show different NMR signals.

Thus, the chemical shift reflects the chemical composition of molecules. As the molecules become more complex, the
15 total number of lines of the absorption spectrum will increase and if we take such a complex molecule as protein then in this case the spectrum will appear as qualitatively shown in Fig. 2.

Something similar will occur also for an aggregate of molecules forming crude oil.

3 Peculiarities of NMR in the Magnetic Field of the Earth

NMR is usually observed in a uniform constant magnetic field, onto which a weak radio-frequency field is superimposed
20 that is perpendicular to the constant magnetic field.

The main requirements of any NMR methods to a source of constant magnetic field are magnetic field stability in time and its high uniformity within the surveyed object.

Due to the fact that the artificially created magnetic field decreases rapidly with increase of distance from magnets, then in order to maintain the constancy of its value within the studied sample the research is carried out in strong magnetic fields
25 of tens of thousands of gauss (Gs). The use of strong magnetic fields for securing NMR entails the need to increase the capacity and frequency of the exciting (sounding) radio signal.

The main difference of surveys in the Earth's magnetic field is very weak field of circa 0.5 Gs. Although a big advantage here is its high stability and uniformity, as well as, undoubtedly, full accessibility and 'free of charge' use of natural magnetic field. In addition, to work in weak magnetic fields do not require high power radio signal

30 The stability and uniformity of magnetic field inside the Earth is particularly important for underground objects including deposits of hydrocarbons, which in terms of NMR are gigantic objects. For example, oil deposit with volume of 1 mln m³ exceeds the ′grain′ of survey in MRI by 10^{15} times, therefore is it impossible to create a stable uniform magnetic field within it.

It should be noted that to date there is a significant number of publications on the NMR method in the Earth's field.
35 Among the first ones are works by M. Packard and R. Varian (Packard and Varian,1953; Packard and Varian,1954). A number of publications in this direction have been summarized in monographs [Derome,1992; Pukhliy and Kovalyov,2010].

A particularly important example of practical implementation of NMR in geophysics are the so-called Magnetic Resonance Sounding systems (MRS system) – systems of search of underground aquifers with the help of NMR.

Such systems operate at relatively shallow depths, up to 100 - 150 m and have not yet been adapted to identify
40 hydrocarbons occurring at much greater depths.

In this regard, we shall consider the operating principle of MRS systems and identify the factors that limit their applicability to work with hydrocarbons.

4 Operating Principle and Characteristics of MRS Systems

The method of magnetic resonance sounding (MRS) is based on excitation, registration and mathematical processing of signal of nuclear magnetic resonance of protons of free water in the Earth's magnetic field.

A special cable is used as an antenna for excitation and reception of a signal. The cable is place on the ground surface in the shape of a square (NUMIS^{Poly} set by IRIS INSTRUMENTS, fig.3) or circumference («Hydroscope» device by SibAkademTechnologies, fig.4).

Excitation of such ground antennas is carried out by radio-frequency sinusoidal impulses. Then amplitude is measured, as well as transverse (T2) and longitudinal (T1) constituents of signals of proton resonance response of water molecules, which gives information about the possible percentage of water content and average pore size.

Pulse power of MRS transmitter is enormous. For example, in NUMIS^{Poly} device impulse voltage of radiofrequency signal reaches 4000 V at current of 600 A. At the same time sounding depth does not exceed 150 m.

It is explained by the fact that MRS systems use extremely inefficient antennas belonging to the type of wire low-hanging loop antennas. The main radiation of such antennas is directed to the zenith, but not in the direction of the object. Directionality of such antennas is shown in fig.5.

In radio such antennas are used for ionosphere communication in accordance with a maximum energy radiation in the vertical plane. Down, under the ground, low-hanging antennas do not radiate (G_{ant} = - 60 dB) and the transmitter power is wasted.

A similar pattern can be observed when the same antennas are used for receiving signals: useful signals are perceived poorly, but atmospheric interference successfully enters the input of the receiver.

5 The main problems of deep detection of hydrocarbons using NMR and their solutions

The first question that arises from experts while getting acquainted with this subject is the following: what power should the transmitter of radio signals have for resonance excitation of hydrocarbons, which deposits are located at a depth of, for example, 5 km.

Let us analyze the underground energetics of radio path.

Path of underground sounding of hydrocarbon deposits consists of two areas:

- ground surface – surveyed object,

- surveyed object – ground surface.

Radio transmitting equipment (RTE) is 'responsible' for the first area (fig. 6) and radio receiving equipment (RRE) – for the second one (fig. 7).

RTE's objective is to create an electromagnetic field concentrated in the direction of the object. The effectiveness of this task implementation is determined by the energy flux density produced by RTE and directed toward the object per unit of time (module of Poynting vector) (Salamatin and Afonin,2008):

$$|S| = P_{trans}\eta_A G_{ant} / S_A, \qquad (1)$$

where: P_{trans} is transmitter power,

η_A – coefficient of efficiency of antenna feeder,

G_{ant} – coefficient of antenna power gain.

S_A - effective antenna area,

Figuratively speaking, the Poynting vector shows where and how much energy is transferred by electromagnetic wave.

We shall estimate the energy efficiency of MRS systems. For this we shall use the system parameters of NUMIS[Poly]:

- transmitter parameters are U_{trans}= 4000 V, I_{trans} = 600 A.

We shall assume feeder efficiency for calculation simplification as η_A = 1.

Coefficient of antenna power gain in vertical downward direction in accordance with fig. 5 is -60 dB, i.e. 10^{-6}.

5   Effective area of loop antenna S_A in accordance with (Markov and Sazonov,1975) is equal to $(0.7 – 0.9)L^2$, where L = 150 m is a side of rectangular loop antenna. We shall set the average value of $S_A = 0.8\,L^2$.

By substituting these values in (1), we obtain the efficiency of the system E_1:

$$E_1= |S|_1 = 4000×600×10^{-6}/0.8×150^2 = 0.133×10^{-3}\ Wm^{-2}. \qquad (2)$$

Thus, to compensate for the extremely low efficiency of low-hanging loop antenna it is necessary to use a very powerful

10   transmitter of radioimpulse signals.

About the same pattern is observed on the receiving path. The same loop antenna used for reception weakens the useful signal and intensely receives noise from the radio.

Altogether despite the immense impulse power of the transmitter, as well as the use of receivers with low own noise, survey depth does not exceed h_1 = 150 m. We shall now determine the power density of signals E_2, which should develop

15   RTE for sounding at depths of up to 5 km.

We shall use for this the obtained MRS system characteristics and extrapolate them to calculate the required efficiency of RTE:

$$E_2 = f(h_1,h_2)·E_1, \qquad (3)$$

where $f(h_1,h_2)$ is function from the depth of MRS sounding and required sounding depths of hydrocarbons h_2.

20   To construct the function $f(h_1,h_2)$ we shall take into account the effects of scattering and absorption of radio waves in the propagation medium (Salamatin and Afonin,2008), and mainly:

- power of electromagnetic radiation in free space decreases inversely as the distance square, i.e. Ps ~ $1/h_2^2$;

- loss in the medium increases proportionally to the distance, i.e. Ps ~ $1/h_2$.

As a result we receive signal loss Ps ~ $1/h_2^3$ and $f(h_1, h_2) = (h_2 / h_1)^3$.

25   Thus, in order to sound an area at a depth of up to h_2 = 5 km we need to ensure that the power density of the signal emitted in the direction of underground objects is equal to

$$E_2 = f(h_1, h_2)·E_1 = (h_2 / h_1)^3 · E_1 = (5000/150)^3 · 0.133·10^{-3} = 4.9\ Wm^{-2}. \qquad (4)$$

This is small value of radiofrequency signal power density. However, it is impossible to achieve such a result the traditional way using inefficient antennas since the transmitter should become unimaginably large at the same time.

30   It is clear that the solution of this task is the development and application of superdirective antennas with a small aperture and high gain coefficient.

Here is an analogy: let us imagine that a dark room is illuminated with a weak one Watt light bulb. It is clear in this case we will not be able to distinguish the corners of the room. If we put this light bulb in a flashlight we will then be able to examine any corner of the room in detail by successively directing the light beam in the desired direction.

35   Thus, the main question of the practical implementation of systems of deep sounding of hydrocarbon deposits passes into the plane of design and engineering development of effective superdirective antennas with a small aperture.

Our way - is the application of super-directive antennas, focusing a relatively small transmitter power in the direction of the object.

The second question that arises during formation of research methods is the choice of ways to increase the reliability of

40   NMR hydrocarbon exploration.

Resonance absorption of hydrocarbon molecules will be very rich as a result of chemical shift. This is especially striking for crude oil. In these cases resonance excitation by a simple sinusoidal signal, as it is done in MRS system, is not sufficient to identify the object.

It is obvious that in order to achieve the maximum degree of identification of the sought-for mineral it is required to excite resonance in all kinds of molecules of the sought-for substance. Otherwise when part of molecules of the mineral does not resonate, the identification accuracy is reduced.

Thus, the main idea of the innovative method lies in "Point-by-point sounding of an area with frequency spectra that excites resonance in the sought-for substance".

Sounding radio-frequency radiation should be highly directional in order to concentrate the transmitter's power in the right direction.

Point-by-point resonance location sounding allows to search for deposits, obtain their ground contours, receive longitudinal and transverse sections, select optimal drilling points. On the basis of these data expected reserves of the deposit are calculated.

6 Variant of Practical Implementation

The magnetic field of the Earth is used as the source of a constant magnetic field to create NMR conditions in the molecules of a sought-for substance at depths of up to 5 km. The spectrum of resonance absorption signal of each of the sought-for minerals is pre-recorded on special test plates (fig.8):

The advantages of using such plates is that the absorption spectra of any minerals, including oil, consisting of hundreds of types of molecules may be transferred to them.

Test plates are further used for transmitter modulation during survey of the corresponding mineral (PatentUA# 86168,2013; PatentUA#86169,2013; PatentUA#86497,2013; PCT/UA2011/000033, 2011; PCT/UA2013/000036,2013).

Deposit survey is carried out with the help of method of pointwise measurements along the deposit's contour as well as along its sections (fig.9):

Sequence of measurement of deposit parameters is shown in Fig. 10.

In measurement point the signal modulated by test plate is directed towards the deposit at α angle. Thanks to chemical shift the spectrum signal is weakly absorbed by various substances in the propagation path, but undergoes significant absorption by oil molecules.

Operator moves along the measuring ribbon with receiver. Response signal is registered at distance from ℓ1 to ℓ2.

Occurrence depths of a horizon are calculated with the help of the following formulae:

$$h1 = \ell1 \cdot tg\ \alpha\ ;\quad h2 = \ell2 \cdot tg\ \alpha \tag{5}$$

Horizon thickness Δh is calculated the following way:

$$\Delta h = h2 - h1 = (\ell2 - \ell1) \cdot tg\ \alpha \tag{6}$$

Similarly, for the following horizons of deposit.

Thus, in each measuring point we receive the following data:

- presence or absence of the sought-for material in the measuring point,

- number of horizons in deposit,

- approximate occurrence depths of horizons;

- approximate horizon thickness.

In some ways these data have something in common with those of exploratory drilling, but geophysical survey of deposits is conducted faster and cheaper.

Test plates with records of resonance frequencies of natural gas recorded at different pressure are placed consecutively in the transmitter modulator during survey of oil deposits. This enables to further define:

- presence of gas caps in the horizons,

- thickness of gas caps,

- pressure in gas caps.

In a number of cases, type of reservoirs and their cap rocks are identified by available samples [Kovalyov et al.,2010; Kovalyov et al.,2011).

In total 40-50 point measuring points are required during work on location for deposit contouring and 20-30 measuring points for constructing sections.

The duration of measurements in each survey point is 30 - 40 min.

7 Examples of Executed Works

1. Testing and practical demonstration of innovative technology were conducted in 2009 on territory of Utah state. The total area is 3600 sq. km. Directly on locality were inspected 5 beforehand unknown for us underground objects, being drillholes and oil-extracting settings.

The following was determined: presence of oil deposits, number of horizons in them, occurrence depth of horizons and their thickness.

Results: efficiency – 100 %, accuracy of depths detection ≥ 98 %.

Fig. 11 is a "Final Report of" on the results of the testing

2. Accidental discharges of 'coalmine methane' occurred at the Zasyad'ko coal mine in Ukraine. In 2010 work was carried out on remote determination of methane origins under the mine shafts.

Drilling results in the point indicated by us confirmed the presence of the alleged source of natural gas and showed high match of our data and drilling data on the number of horizons, their occurrence depth as well as gas pressure in the horizons (Table 1).

Results: efficiency – 100 %, accuracy of depths detection ≥ 95 %.

"Final report" on the results of drilling is shown in Fig.12.

8 Conclusion

The article considers theoretical possibilities and applied questions of NMR effect use with the aim of search and prospecting of hydrocarbon deposits.

It is shown that the main idea of the innovative method lies in "Point-by-point sounding of an area with frequency spectra that excites resonance in the sought-for substance". Using a highly directional antenna at the same time is compulsory.

Sequence of operations of ground sounding of hydrocarbon deposits was developed as well as calculating procedure of occurrence depth of horizons and their thickness.

Works conducted on the basis of the considered method have shown high efficiency, accuracy and results repetition.

This initial work was aimed at creating a summary about the unique capabilities of an innovative NMR technology to carry out a direct detection of hydrocarbons, minerals and water deposits to a depth of 5 km. without interpretation by highly directional antenna. The use of these features can significantly reduce an exploration budgets in exploration geophysics.

9 Data availability

The authors come out with the statement that all the underlying data in the article are publicly available and are used by Copernicus Publications on their discretion.

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Yurchuk A. Technical Director of the Institute, the acquisition of field data for the article; the technical design of the article.

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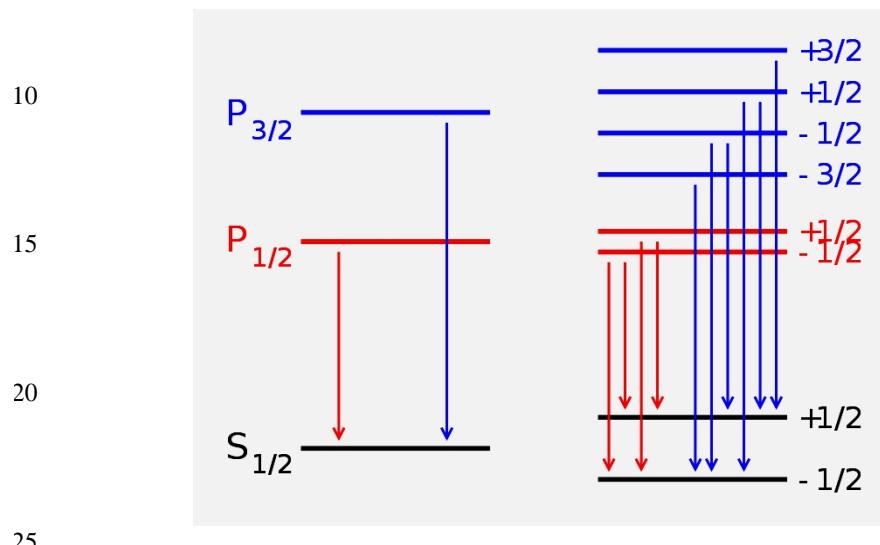
Figure 1: Example of the Zeeman effect for alpha transition in Lyman series
On the left are unperturbed levels. On the right are levels split under the impact of magnetic field. The arrows indicate dipole
allowed transitions.
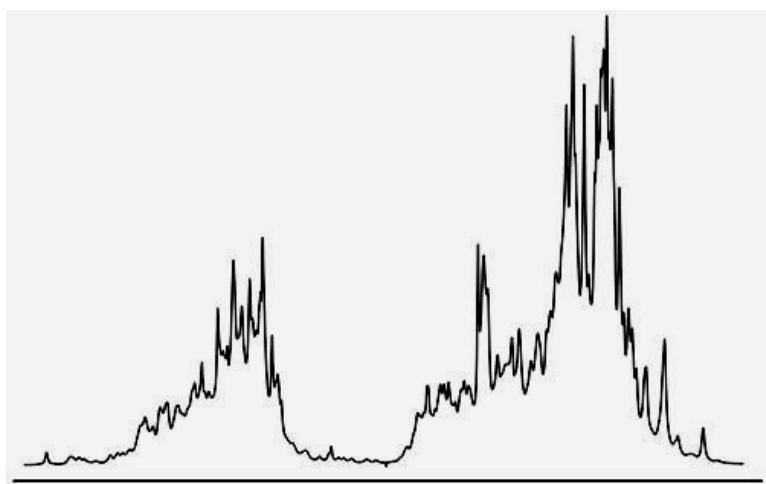
Figure 2: NMR absorption spectrum by protein molecule
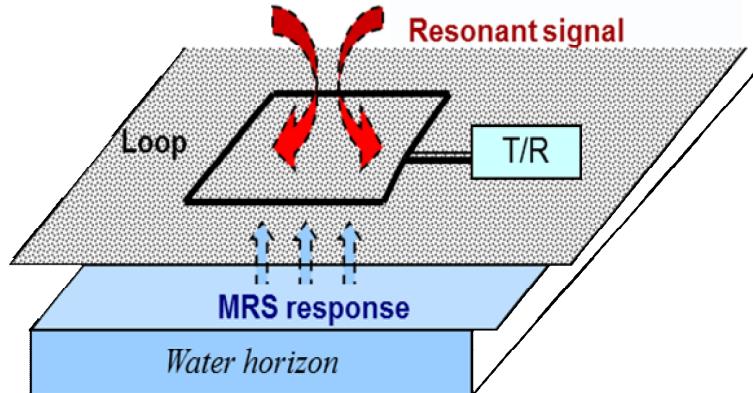
Figure 3: Arrangement on location of MRS equipment of IRIS INSTRUMENTS
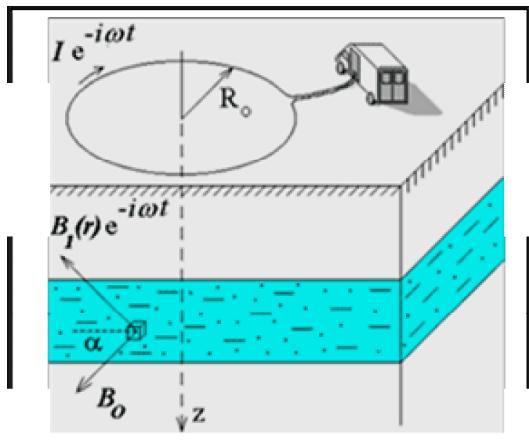
Figure 4: Arrangement on location of MRS equipment "Hydroscope"

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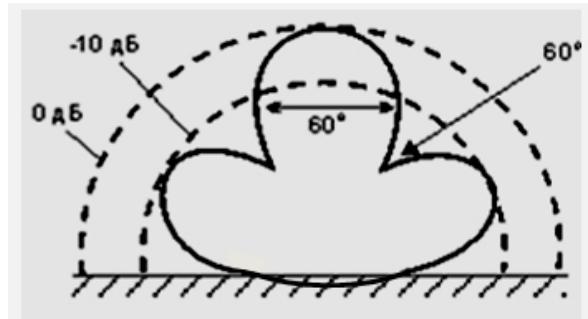
Figure 5: Directionality of low-hanging horizontal loop antenna

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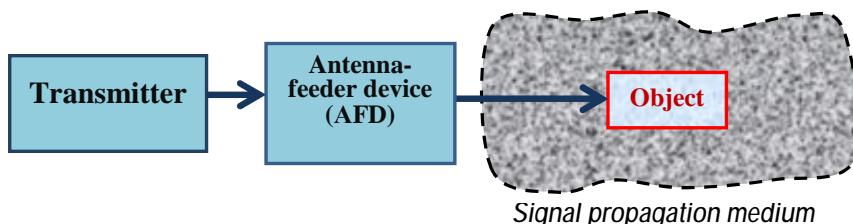
Signal propagation medium

30   **Figure 6: Radio transmitting equipment**

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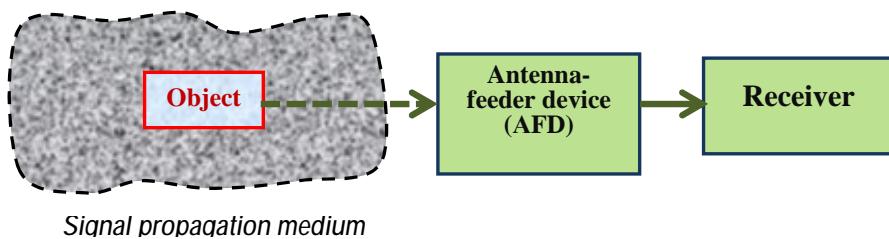
Signal propagation medium

45   **Figure 7: Radio receiving equipment**

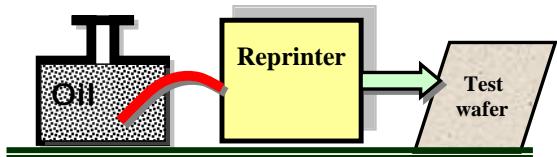
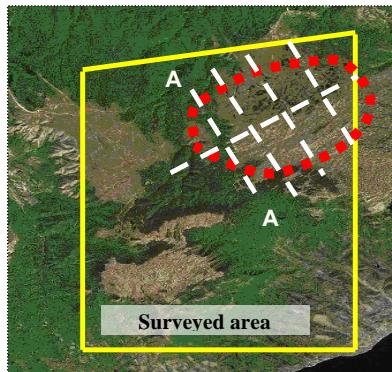



Figure 8: Diagram of recording of spectral characteristics of oil onto the test plate

10   **Figure 9: Diagram of point-by-point deposit survey**

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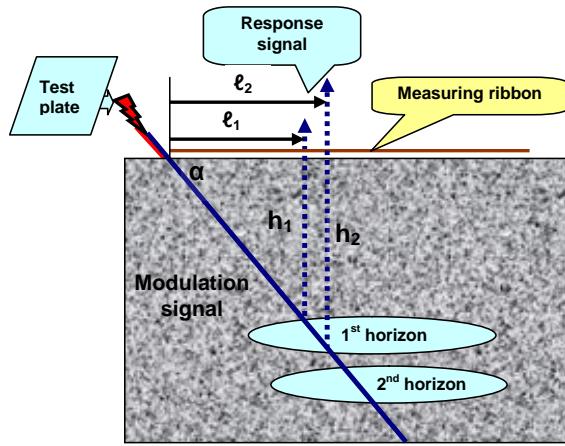
Figure 10: Diagram for measuring the parameters of deposit occurrence

Figure 11: "Final report" on the results of the testing

Figure 12: "Final report" on the results of drilling

TABLE 1:

Number of horizon	Depth,m our data/drilling	Gas pressure, kg/sq.cm. our data/drilling
1	544-583/535-595	10- 20/16
2	973-1043/906-1020	15-20/92*
3	1272-1317/1266-1324	18-20/_ **
4	1753-1857/1794-1808	150-160/164

Parameters of the survey/drilling

*Gas flow rate of 0.26 cubic meters per day
**The drilling fluid disappeared from cavity