Spin Noise Spectroscopy

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Abstract. Development of Spin Noise Spectroscopy (SNS) beginning from the first experiment on detecting magnetic resonance in the Faraday-rotation noise spectrum in sodium vapor up to its recent achievements is briefly reviewed.

1. Introduction
Spin noise spectroscopy (SNS) is now considered as a new branch of magnetic resonance spectroscopy based on detecting spontaneous fluctuations of magnetization by means of Faraday-rotation technique. The first experiment that demonstrated feasibility of such an approach was performed in 1981 [1]. For us, the story of what is now called ‘spin noise spectroscopy’ has begun in the middle of seventies when we started working on achieving the limiting (shot-noise-limited) polarimetric sensitivity. In the experiment [1], we detected spontaneous noise of transverse magnetization of sodium atoms in a magnetic field at Larmor frequency. In this communication, we briefly consider the reasoning underlying this first experiment, review the conceptual and methodological aspects of this experimental approach, and discuss some possible ways of its further development.
Fig. 1. Formation of “paramagnetic” part of the Faraday rotation. A difference in populations of the ground-state sublevels creates difference in optical absorption for two circular polarizations, which (in conformity with Kramers-Kronig relations) makes different refractive indices for two circular polarizations in the transparency region near the absorption line. This ‘magnetic circular birefringence’ is revealed as rotation of the polarization plane (Faraday rotation). Magnetic splitting of the levels (giving rise to the ‘diamagnetic’ magnetooptical effects) is ignored.

2. Conceptual background

In 70s, when the experiment [1] was planned, all the fundamental information needed to predict its feasibility and to evaluate the magnitude of the effect was available. Information capacity of the noise spectroscopy seemed to be, in principle, clear. Correlation characteristics of the ‘noise signal’ and its spectrum were known to contain information about dynamics of the process underlying the observed signal, and, in conformity with the fluctuation-dissipation theorem, to be closely related to the appropriate susceptibility spectrum of the system. Some experiments of this kind, demonstrating potentialities of the noise technique, have been already fulfilled [2]. As for the magnetization noise, the fundamental possibility of its observation in the nuclear spin systems was mentioned by Bloch in 1946 in its classical monograph “Nuclear Induction”. Of course, it was not essential that Bloch spoke about nuclear, rather than electronic, spins. The more so, that possibility of observation of regular signals of coherent spin precession in atomic system by means of magneto-optics was also well justified in experiments on optical pumping [3]. So, the question was of more practical, rather than fundamental, nature: it was not clear whether it is possible experimentally to detect the spin noise of essentially macroscopic systems for reasonable times with sufficiently high signal-to-noise ratio and, after all, whether it has any sense.

Fig. 2. Measuring spin noise in two configurations: (a) in a longitudinal magnetic field (Faraday configuration) and (b) in a transverse magnetic field (Voigt configuration).
At that time, we were well familiar with magneto-optics of atomic and solid-state paramagnets and knew that the Faraday rotation (or, more exactly, its “paramagnetic” part) is proportional to magnetization of the spin-system and can be used for magnetic measurements. So, by measuring fluctuations of the Faraday rotation, we could, in principle, measure fluctuations of magnetization of the system or what is called ‘spin noise’.

3. Experimental geometries
The first idea was to measure fluctuations of longitudinal magnetization (Fig. 2a), which may, in principle, give important information about longitudinal relaxation time $T_1$ and about its dependence on temperature, magnetic field and other external perturbations. Information of this kind was usually obtained from the EPR data. This experimental approach was, in fact, close to the paramagnetic relaxation technique employed by the Dutch physicist Gorter, before the advent of magnetic resonance, for measuring magnetic susceptibility of paramagnets in parallel fields [4]. The essential difference between these two approaches was that in our case the system was not supposed to be perturbed by the external oscillating field.

After a short discussion, however, we came to conclusion that this idea was not good, at least, from the methodical viewpoint, because it implied measuring the “noise signal” at low frequencies where the measurements are usually hampered by many other sources of noises, and the peak of the noise power density at zero frequency should be observed anyway. So, more reasonable was to measure transverse fluctuations of magnetization. In this geometry (the so-called Voigt configuration, Fig. 2b), a random transverse fluctuation of the magnetization should precess around the magnetic field during a time interval of the order of $T_2$ (dephasing time or transverse relaxation time) and then be replaced by another random realization of the transverse magnetization. The linearly polarized probe light, in this geometry, should experience preferential oscillations of the polarization plane azimuth at the frequency of Larmor precession of the spin. In other words, in this configuration, we expected to observe a peak, in the Faraday rotation noise spectrum, at the frequency of magnetic resonance and, thus, to obtain the magnetic resonance spectrum of the unperturbed system.

4. Problems of sensitivity
The main requirements to experimental arrangement for such measurements, dictated by the necessity to achieve the greatest polarimetric sensitivity, implied

1) maximizing the probe beam intensity (to reduce relative level of the shot noise, proportional to $N^{-1/2}$, where $N$ is the number of photons);
2) minimizing the level of excess light-intensity noise;
3) minimizing the total number of spins contributing to the Faraday rotation (to increase relative value of the fluctuation). This requirement strongly differs from what is usually needed in measurements of susceptibility, when signal increases with the size of the system;
4) maximizing specific Faraday rotation (per spin);
5) maximizing total Faraday rotation for the completely aligned spin system.

The last item follows from the fact that the “noise signal” proportional to $n^{-1/2}$ ($n$ is the number of spins) is calculated as a relevant fraction of total rotation, i.e., of the rotation of completely oriented spin-system. One can easily see that the total rotation may substantially differ for samples with the same number of spins ($n$), but with different shape (Fig.3). Therefore, to maximize the “noise signal” for a fixed number of spins, the sample volume probed by the light should be maximized in length and minimized in cross section or, in other words, the light beam passing through the sample should be tightly focused.
It is noteworthy that, at that stage, we seriously discussed the possibility of using thermal source (incandescent lamp) for detecting these small polarization fluctuations. The point was that the intensity fluctuations of laser sources, at that time, exceeded the shot noise level by 4 - 6 orders of magnitude, and, as a result, the laser sources, with all their unquestionable merits, seemed unsuitable for ultimately sensitive polarimetric measurements. Still, these two types of the light sources were so incomparable in their brightness, that we stopped at lasers.

The problem of excess noise suppression for laser sources was solved using several expedients [5], the most substantial of them being balanced detector.

5. Balanced detector

The detection unit of the polarimetric system consisted of a polarization beamsplitter with its two outputs coupled to two photodetectors included into a differential circuit so that their photocurrents were subtracted at the load resistor (Fig. 4). When the plane of polarization of the incident light made an angle of $45^\circ$ with polarizing directions of the beamsplitter, photocurrents of the two photodetectors were cancelled at the load resistor, while the signals resulted from oscillations of the incident beam polarization plane rotation, with opposite phases on the detectors, were summed up. In this way, we have managed to suppress the excess noise by three orders of magnitude and to achieve the shot-noise limit of polarimetric sensitivity lying, for the laser power of several hundreds mW, in the range of $10^{-8}$ rad (see [5] and references therein). In those experiments, the balanced detector, which is now produced commercially, was applied to polarimetric measurements, to the best of our knowledge, for the first time.
6. The first experiment

The experiment on detection of magnetic resonance in the Faraday rotation noise spectrum [1] was performed on sodium atoms at a pressure of ~ 10 Torr. Schematic of the experimental setup is shown in Fig. 5. As a light source, we used a cw dye laser tuned to close vicinity of the D1 or D2 line of Na. The cell with sodium vapor was placed into a transverse magnetic field created by a system of Helmholtz coils. The field was slightly modulated at a frequency of ~ 10^2 Hz. The output signal of the balanced detector was selectively amplified at a frequency of 1.3 MHz, then quadratically detected, lock-in amplified at the frequency of field modulation, and recorded as a function of the applied magnetic field. Figure 6 shows two typical plots of the spin-noise resonance recorded with a time constant of 2 s. The probe beam was tuned to the edge of D1 line or to the midpoint between the two lines (for more details, see Ref. [1]).
7. Perturbative or not?
One of the questions that arose after successful realization of the experiment was whether this technique could be considered nonperturbative or not. At first glance, it looked absolutely passive and nonperturbative because it was based on detecting Faraday rotation in the transparency region, and the probe beam, as it seemed, did not touch, in any way, the atoms under study. But, on the other hand, it could not be nonperturbative for evident reasons: The initially monochromatic light, after passing through the spin system, becomes modulated. Therefore, it acquires sidebands with shifted frequencies. It means that some photons of the beam experience inelastic scattering, which implies energy exchange between the light and spin-system. To resolve this contradiction, we addressed our famous theorist V. Perel, who immediately accepted this problem for consideration and very soon, in coauthorship with Gorbovitskii, has published the paper [11], where it has been shown that the noise of the Faraday rotation at the frequency of Larmor precession can be considered as a result of coherent forward Raman scattering of the incident laser beam. So, this process can be regarded, to a considerable extent, nonperturbative, because the probe light interacts with macroscopic fluctuations of the system, and does not select any particular spins to flip. The technique becomes more and more perturbative as we pass from macroscopic ensembles to microscopic ones.

8. Nuclear spin noise
Soon after our work on detection of spin-noise resonance in atomic vapor, an ideologically the same experiment has been performed on a nuclear system [7]. As a spin system, in this experiment, was used an ensemble of nuclei $^{35}\text{Cl}$ in a NaClO$_3$ crystal. The experiment was performed at 4 K, and the magnetization noise was detected by means of SQUID. This was the first experiment that demonstrated resonant spin noise predicted by F. Bloch on a nuclear system. Here, the number of spins contributing to the signal was much larger than in our experiment with electron spins, and the sensitivity of the detection system was much lower. As a result, the peak of spin noise at the frequency of nuclear quadrupole resonance of $^{35}\text{Cl}$ was detected with the signal-to-noise ratio of a few units for the accumulation time of 7 hrs. Still, further technical advancement in this area made it possible to observe the nuclear spin noise at room temperature [8, 9] and even to develop, on its basis, the method of nuclear spin noise imaging [10].

9. Primary development of SNS
For a decade or so, our publication remained practically unnoticed. The Faraday-rotation noise spectroscopy as a new technique of ESR detection was appreciated at the end of last century, first of all, by a group of researchers from Los Alamos National Laboratory [11], who reproduced the experiments with alkali atoms and announced the spin noise spectroscopy as an efficient probe of spin dynamics and magnetic resonance. In [12], it was shown, in particular, that magnetic resonance in the Faraday rotation noise spectrum can be also detected in the longitudinal magnetic field. This may occur for the ESR transitions allowed in the ac magnetic field oscillating along the dc magnetic field. This result can be easily comprehended in the context of the fluctuation-dissipation theorem, which establishes direct relationship between the spectra of susceptibility and fluctuations (in this case, both with respect to longitudinal magnetization).

A highly important step in development of SNS was made by Oestreich et al. [13] who have managed to successfully apply this technique to semiconductors. At this stage, these experiments demonstrated more a fundamental applicability of this experimental approach to semiconductors rather than its practical importance, because the signal became visible after its accumulation for 20 hrs.
10. A breakthrough in SNS – FFT spectrum analyzer
A real breakthrough in this field of research has happened when, in the system of data acquisition, the sweeping spectrum analyzer was replaced by the one with the real-time fast Fourier transform processing system [14]. The conventional sweeping spectrum analyzer, as is known, measures the noise only in a very narrow (resolved) frequency interval at a time, thus disregarding most of the information coming to the photodetectors. The FFT spectrum analyzer, on the contrary, digitizes the signal in the whole operation bandwidth of the system (with a sampling rate of around $10^9$), performs FFT of the signal in real time, and averages the spectrum thus obtained. As a result, the whole bulk of information contained in the signal is being processed and stored.

This technical advance allowed one to increase sensitivity of SNS by a few orders of magnitude and made it suitable, in particular, for studying spin dynamics in low-dimensional semiconductor systems (quantum wells, quantum dots, etc.) highly important for the present-day technological applications.

11. GHZ and THZ spin noise spectroscopy
An interesting idea was proposed in [15] to overcome limitations associated with the bandwidth of optical detectors. For that purpose, it was proposed to use a pulsed laser with a high repetition rate instead of a cw laser, as a source of probe light. In this case, we can observe, in the spectrum of the detected signal, not only the peak at the frequency of magnetic resonance $f_R$, but also the peaks at the frequencies shifted from $f_R$ towards lower frequency by multiples of the repetition rate $f_0$. As a result, the frequency of the spin noise resonance under study can be transferred to the frequency range $[0, f_0]$. This experimental approach allowed the authors of [15] to detect spin noise at Larmor frequencies up to 16 GHz. Under these conditions, the total bandwidth of the spectrum analyzer and of the photodetectors may not exceed the repetition rate $f_0$. At the same time, the measured spin dephasing times are limited, in this technique, by approximately half of the repetition rate. It is important that application of this technique does not reduce sensitivity of SNS as compared to its conventional version.

Another remarkable approach to solution of the problem of widening of the bandwidth of the Faraday-rotation-based SNS was proposed in [16]. The authors of this idea proposed to use, for this purpose, an extremely broad bandwidth of optical femtosecond pulses and to measure correlation characteristics of the magnetization of the medium with a very high time resolution. For that purpose, the probe beam was proposed to form from pairs of ultrashort femtosecond pulses (not resolved by the detection system) and to measure the Faraday rotation noise power as a function of time delay between the pulses. This technique allows one to directly detect decay of the transverse magnetization in time domain and to expand the accessible frequency range up to several terahertz.

12. Cavity enhancement of spin noise
In [17], it was shown that the Fabry-Perot cavity may be highly sensitive to modulation of its intracavity anisotropy at frequencies multiple of its intermode spacing. This effect can be easily understood in terms of light pulse traveling over the cavity back and forth and has much in common with the mode-locking effect in lasers. Figure 8 shows frequency dependence of the polarization modulation gain factor for different positions of the anisotropic element inside the cavity (for more detail, see [17]). This effect can evidently be used in SNS for selective amplification of the polarization modulation signal. Amplification, in this case, is performed in optical (rather than electronic) channel, with the gain factor being approximately equal to Q-value of the cavity ($\sim 10^2 – 10^3$).
More interesting, however, is to use this effect for creating an all-optical spin-noise spectrometer [17]. The idea of this proposal is as follows (Fig. 8). Let the sample under study be placed inside an optical cavity. A linearly polarized laser beam incident on the cavity hits one of its longitudinal modes and thus passes through the cavity with no loss. At the exit of the cavity, we place a linear polarizer crossed with the polarizer at the entrance, so that no light can pass through the system. Now, if we start sweeping the transverse magnetic field applied to the sample, then, at the moment when Larmor precession frequency of the spins becomes equal to (or multiple of) the intermode frequency of the cavity, oscillations of the polarization plane at the exit of the cavity appear to be strongly enhanced, and the light starts to pass through the output polarizer. In this case, it does not matter how high is the frequency of these oscillations. We detect only dc component of the transmitted light (or modulated light intensity if the transverse field is modulated). A unique property of this arrangement is that it allows one to detect magnetic resonance in the Faraday rotation noise spectrum at any frequency without any broadband electronics and broadband photodetectors. What we observe is just a result of mutual resonance of the spin system and Fabry-Perot cavity. A serious drawback of this proposal is that the transmitted light intensity, at small rotation angles, is quadratic in angle and, for small angles of rotation, becomes negligibly small.

13. Conclusions
At present, due to a number of technical advances, spin noise spectroscopy turns into a standard and fairly unique tool of magneto-resonance investigations. With the advent of FFT spectrum analyzers into arsenal of technical means of SNS, a considerable portion of labor has been reload on the shoulders of electronics, and the information of a rather sophisticated nature became fairly chip. The absence of any HF oscillators allows one not only to avoid perturbation of the system, but also to substantially simplify technical structure of the setup and to make it much more convenient in handling. High sensitivity of this technique in combination with high spatial resolution makes it unique for studying spin systems in micro- and nanostructures. A sort of 3D spin-noise tomography has been demonstrated in [18], where the medium was probed by the waist of a tightly focused laser beam, which provided the main amount of the noise signal. In [19], it was shown that the time-of-flight broadening may be highly important in SNS of semiconductor materials. Spin noise spectroscopy is now successfully applied for studying electron statistics and evaluating the degree of electron localization [20, 21].

The main research on SNS is now concentrated around semiconductor systems of different morphology, whose spin-related properties are considered to be highly promising for applications in photonics and information science. Still, SNS also finds application in studying, along with diluted paramagnets, spin glasses and ferromagnetic fluids. As a nonperturbative technique, it is considered highly efficient in studies of ultracold atoms. In spite of close connection between the fluctuation and
susceptibility spectra, formulated by the fluctuation-dissipation theorem, their information capacities are not identical. In many cases it may appear important that the measurements of susceptibility imply coherent perturbation of the system, while fluctuations are caused by its random incoherent thermal motion.

We did not mean, by any means, to give here a comprehensive review of this topic. We just tried to show that the idea of detecting magnetic resonance in the Faraday-rotation noise spectrum, that looked, at the beginning, as an academic trick, useful primarily for tutorial purposes, turned into a highly efficient practical tool of magneto-spectroscopic research.

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CORRIGENDUM

On page 7, the third sentence of section 12 should be amended to read: We have calculated the frequency dependence of the polarization modulation gain factor for different positions of the anisotropic element inside the cavity (for more detail, see [17]).

On page 8, 2 lines below figure 7 the reference to the figure should be changed to (Fig. 7).