Optical and RF Spectroscopy of Spin Noise

E B Aleksandrov 1 and V. S. Zapasskii 2
1 Ioffe Physico-Technical Institute, Saint-Petersburg, Russia
2 Saint-Petersburg State University, Spin Optics Laboratory, Saint-Petersburg, Russia

E-mail: zap@vz4943.spb.edu

Abstract. In the lecture, we briefly outline basic ideas underlying the method of spin noise spectroscopy and consider informative capabilities of the optical and RF spectroscopy of spin noise power detected by means of magneto-optics.

1. Introduction
The method of spin noise spectroscopy (SNS) proposed in early 80s [1] and substantially developed during the last decade [2, 3] is considered primarily as a specific optical technique of magnetic resonance. Spectral shape of the RF “noise signal”, in the vicinity of the Larmor frequency of the spin-system, is determined by its magnetic characteristics and, in fact, provides magnetic resonance spectrum of the system. Behavior of the other degree of freedom of the Faraday-rotation-based SNS, related to its optical channel, is controlled not only by magneto-optical characteristics of the system, but also, in a rather sophisticated way, by interplay of optical transitions associated with different spin-systems of the paramagnet. [4].

2. Basics of the SNS
The basic idea of the spin noise spectroscopy (SNS) is very simple. Magnetization of a spin-system in an external magnetic field is known to be the result of summation of a multitude of projection of magnetic moments, each of them being a random quantity. The equilibrium nonzero magnetization is created due to the difference in probabilities for each particular spin to be oriented parallel or anti-parallel to the applied field. As a result, the field dependence of the equilibrium magnetization of the spin-system is described by the Brillouin-type function, and, for an isotropic paramagnet, the spin-system magnetization, in a fixed magnetic field, has a definite fixed value and is directed exactly along the applied field.

This is correct, however, only to within magnetization fluctuations, which are inevitable for a paramagnet with a finite number of spins. If we look at the tip of the magnetization, so to say, through a microscope, we will see that it experiences fluctuations both in magnitude and in direction. These two types of magnetization fluctuations – along and across the applied magnetic field – appear to be essentially different (Fig. 1).
correlation time of the longitudinal magnetization fluctuations is controlled by the so-called longitudinal relaxation time of the spin-system \((T_1)\), so that the correlation function of the process has an exponential shape with the decay constant \(\sim T_1\). In conformity with the Wiener-Khinchin theorem, spectrum of this magnetization noise, represented by the Fourier transform of the correlation function, is shaped as a Lorentzian with the width \(\sim T_1^{-1}\) centered at zero frequency.

An essentially different behavior is revealed by fluctuations of the transverse magnetization. Any random nonzero transverse component of the magnetization will evidently exhibit Larmor precession around the applied magnetic field. This precession will last during the time of about \(T_2\) (dephasing time or transverse relaxation time) and then will be replaced by another realization of transverse random component with different magnitude and phase. As a result, the transverse magnetization has to reveal an excess noise at the frequency of Larmor precession. Correspondingly, the correlation function of this process will have an oscillatory behavior with the decay time \(\sim T_2\), while its spectrum will be represented by a Lorentzian with the width \(T_2^{-1}\) centered at Larmor frequency of the spin-system (Fig.1). In other words, the transverse magnetization noise reveals the spectrum which we usually refer to as the magnetic resonance (EPR) spectrum. So, the paramagnet in an external magnetic field, under equilibrium conditions, should reveal its EPR spectrum without any external perturbation.

All this looks good on paper. In reality, for a macroscopic paramagnet, these thermodynamic fluctuations appear to be extremely small, and the experimental problem of their detection may seem, at first sight, hardly solvable. Fortunately, there exist polarimetric methods of magneto-optics that allow one to detect tiny variation of magnetization of a paramagnet practically with no perturbation of the spin-system.

This experimental technique is based on the fact that the magnetic circular anisotropy of a paramagnet (circular birefringence, responsible for the Faraday rotation, and circular dichroism, responsible of the field-induced ellipticity) contains the so-called “paramagnetic” contribution which is directly proportional to spin-system magnetization [5]. The most convenient method of optical detection of magnetization implies the use of the Faraday rotation (FR), which can be observed in the region of transparency where the spin-system can be probed in a perturbation-free way. Sensitivity of the FR technique to small variations of the magnetization is determined not only by the proportionality factor controlling conversion of magnetization to FR (which depends on the value of spin-orbit interaction in the system and on the light wavelength), but also by polarimetric sensitivity of the experimental setup. The possibility of detecting spontaneous noise of magnetization of real macroscopic spin-systems became clear after achieving shot-noise-limited polarimetric sensitivity lying, for the laser power of several tens of mW, in the range of \(10^{-8} - 10^{-9}\) rad (see [6] and references therein).

The first experiment on detection of magnetic resonance in the FR noise spectrum [1] was performed with sodium atoms at a pressure of \(\sim 10\) Torr. Schematic of the experimental setup is shown in Fig. 2. As a light source, we used a cw dye laser tuned to close vicinity of the D₁ or D₂ line of Na. To decrease the number of atoms contributing to the FR noise and thus to increase the level of the detected noise, the laser beam was focused in the cell. Excess noise of the laser source was suppressed by the balanced detector which subtracted correlated intensity fluctuations in two arms of the polarization beamsplitter and
summed up uncorrelated intensity fluctuations related to polarization fluctuations of the incident beam. The cell with sodium vapor was placed into a transverse magnetic field created by a pair of Helmholtz coils. The output signal of the balanced detector was selectively amplified at a frequency of 1.3 MHz, then quadratically detected (rectified), lock-in amplified at the field modulation frequency, and recorded as a function of the applied magnetic field. The signal of spontaneous magnetic resonance was recorded, in these experiments, with the signal-to-noise ratio (a sort of “noise-to-noise” ratio) was about 100:1 for the accumulation time ~ 2 s. This was the first experimental demonstration of feasibility of the spin noise spectroscopy.

Until the beginning of our century, however, the experiment [1] remained more or less unnoticed. A considerable impact to development of the SNS was given by publication of Crooker with coauthors [2], in which the experiment [1] was extended to some other alkali atoms (Rb and K) and by the experiment of Oestrich et al. [7], in which the authors have managed to apply this technique to solid semiconductor systems. A real technical breakthrough occurred in the SNS in 2007 [8], when the sweeping spectrum analyzed, in the system of data acquisition, was replaced by the one with the real-time fast Fourier transform (FFT) processing system. As a result, the sensitivity of the technique was increased by more than two orders of magnitude, and SNS turned into a real practical tool of the magnetic resonance spectroscopy.

![Schematic of the first experiment on detection of magnetic resonance in the FR noise spectrum.](image)

**Fig. 2.** Schematic of the first experiment on detection of magnetic resonance in the FR noise spectrum.

The use of optical field for monitoring magnetic structure of the spin-system gives birth to two spectroscopic aspects of the technique, related, respectively, to the RF spectrum of the spin noise (magnetic resonance spectrum) and to optical spectrum (wavelength dependence) of the spin noise power (Fig. 3). These two aspects of the SNS are its complementary sides which enrich each other and make the technique much more informative. Consider them successively.
3. RF spectroscopy of spin noise
Initially, SNS was considered just as a perturbation-free method of detection of magnetic resonance. In other words, all the information was supposed to be obtained from RF spectrum of the spin noise, with the role of the optical channel being restricted by providing maximum value of the signal. Specific properties of the RF spin-noise resonance, whose lineshape and position obey standard laws of the magnetic resonance spectroscopy, are (i) absence of any ac-field-induced distortion of the resonance line (in view of absence of any ac field) and (ii) independence of the spin noise (magnetic resonance) signal of the population difference between magnetic sublevels of the spin-system (no magnetic polarization of spin-system is needed to detect the spin-resonance signal). In addition, the FR-based SNS reveals the typical for optical methods high special resolution achieved under condition of tight focusing of the probe beam (which is anyway needed to increase sensitivity of the technique). For localized spins, this feature allows one to study relief of inhomogeneity of the sample at high special frequencies and may considerably mitigate requirements to the magnetic field homogeneity. For non localized spins, as, e.g., in atomic vapors or in semiconductors with free carriers, there arises an addition “time-of-flight” broadening, which depends on the light beam cross section and can be used to get information about kinetic characteristics of the spin ensemble. In particular in [9-11], this opportunity was used to distinguish between localized and delocalized conduction-band electrons in n-GaAs and to detect the effect of Brownian motion of the electrons in the conduction band on the spin noise line width.

4. Optical spectroscopy of spin noise
Magnetization of a spin-system is converted into the FR in accordance with the spectral behavior of the latter. This spectrum, in turn, for an isolated absorption band, has a dispersion-like shape as shown in Fig. 4a Accordingly, the FR noise power (integrated over the RF spectrum) is expected to be proportional to the FR (or Verdet constant) squared, vanishing at the center of the band where the FR turns into zero (Fig. 4b). This is, however, not necessarily the case [4].

When an isolated absorption band (or a more complicated optical spectrum) is related to a single spin-system or, in other words, the absorption spectrum of the ensemble coincides with that of a single spin-system, then, indeed, dependence of the spin noise power on optical frequency is described by FR squared (Fig. 4,b), and the behavior of the spin noise spectrum with optical frequency will vary in magnitude as shown in Fig. 4.c. If, however, the absorption spectrum comprises several components related to different spin-systems, the situation changes.
Consider, as an example, an optical spectrum comprised of two closely spaced components (Fig. 5a). The FR spectrum of the system will look as shown in Fig. 5b regardless of whether these components are related to one and the same spin-system or not. At the same time, spectral behavior of the FR noise (spin noise power) will be essentially different for these two cases. While in the case of a single spin-system fluctuations of the FR of the two components appear to be correlates, and the FR noise is described by the total FR squared, in the second case, fluctuations of the FR in two components are uncorrelated, and the optical spectrum of the FR noise will be described by the sum of squared of the two partial contributions of the components. As a result, optical spectra of the spin noise, in these two cases appear to be strongly different (Fig. 5, c, d).

The most spectacular example of this difference is given by the case of inhomogeneously broadened spectral bands, when a great multitude of spectral components comprising the band provide independent contributions to the FR noise. In this case, the FR noise of the inhomogeneously broadened band looks as shown in Fig. 4, d, with no dip in the center of the band, where the FR proper vanishes. As the ratio of inhomogeneous broadening to homogeneous increases, the depth of the dip in the optical spin noise spectrum decreases and, what is highly important, the magnitude of the noise signal increases (Fig. 6). This fact provides additional advantages of the spin noise spectroscopy with respect to inhomogeneously broadened systems. These conclusions have been confirmed experimentally in [4].

**Fig. 4.** Optical spectra of absorption and FR (a), FR squared (b), spin noise resonance for the case of homogeneous (c) and strongly inhomogeneous (d) broadening.

5. Conclusions

The spin noise spectroscopy which is getting more and more popular for studying diverse paramagnets possesses two channels of spectroscopic information, corresponding to the RF and optical frequency range and contained primarily in the lineshape of the RF and optical resonances of spin noise power. These two channels in combination considerably widen experimental potential of this promising tool of the EPR spectroscopy providing it with new sources of information.
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