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A study of TOF-MIEZE reflectometry for nanomagnetic dynamics

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Abstract. Through the combination of neutron reflectometry and modulated intensity by zero effort instrument (MIEZE) technique, it is possible to detect the inelastic and quasi-elastic scattering on the surface and at the interface of thin films. In particular, the combination of the time-of-flight (TOF)-MIEZE technique and polarized neutron reflectometry enables us to study the nanomagnetic spin dynamics in a thin film. We show experimental results of TOF-MIEZE signals of neutrons reflected by the Fe thin layer and Fe/Si multilayer as the feasibility of new technique for investigation of nanomagnetic dynamics.

1. Introduction

Polarized neutron scattering is a very powerful tool for the analysis of magnetic structure and neutron reflectometry is a powerful tool for the analysis of the structure of thin films. Polarized neutron reflectometry (PNR) is the combination of these two techniques, which is used mainly for the investigation of magnetic thin films, magnetic super-lattices, or any kind of magnetic hetero-structure. To date, most PNR work has been conducted with a static or quasi-static sample environment [1]. Here the resolution of time-resolved measurement is limited to order of seconds even in state-of-the-art neutron reflectometer instrument owing to the neutron intensity. Recently frequency-dependence PNR experiments have been conducted to measure magnetization reversal in thin Fe films [2]. The magnetization reversal was observed by measuring polarized neutron specular reflectivity under high-frequency external magnetic field. It is an excellent approach to investigate the effect of interfaces on the magnetization, however, sample applications and the frequency of the external magnetic field are limited. Inelastic neutron scattering (INS) and quasi-elastic neutron scattering (QENS) are powerful tools to study the dynamical structure of materials. If applied to the reflection geometry, in the case of non-magnetic reflection from a thin film, the neutron energy change is limited in the direction perpendicular to the film surface. Here we assume that incoherent scattering is negligible. In order to analyze dynamical structure on the surface and the interface of thin films, it is necessary to conduct the energy analysis of off-specular scattering neutrons. However, off-specular scattering also contains elastic neutron scattering (ES) caused by in-plane structure of a sample coming from interfacial roughness and/or inhomogeneity of composition. The intensity of ES is normally much larger than those of INS and QENS. Hence, we need to extract neutrons undergoing the INS and QENS processes from off-specular signals caused by in-plane structures. In case of magnetic reflection with polarized neutrons, the neutron energy change is not limited in the direction perpendicular to the film surface. Because neutron magnetic scattering processes describe interactions between a vector of neutron magnetic moment and a vector of magnetic potential in a sample. By employing neutron polarization analysis, it is possible to select velocity changed neutrons by magnetic INS and QENS processes. This implies the possibility of a new research method for the in-plane spin dynamics of the magnetic film. Here we point out the feasibility of direct observation of spin relaxation of nanomagnetic system by employing a combination of PNR and the time-of-flight modulated intensity by zero effort (TOF-MIEZE) technique. It is possible to use any magnetic sample environment including frequency-dependence magnetic field. To our knowledge, there have been no previous attempts to measure to measure spin relaxation of the in-plane magnetization up to order of nanoseconds with neutrons. In this paper, we show 200 kHz TOF-MIEZE signals of neutrons reflected by the Fe thin layer and Fe/Si multilayer as a feasible study for the study of nanomagnetic dynamics.

2. Features of the TOF-MIEZE spectrometer

The neutron spin echo (NSE) technique [3, 4, 5] is a unique spectroscopic method to investigate the slow dynamics of molecules and molecular assemblies. It directly measures the intermediate scattering function $S(Q, \tau)$, where Q and τ are the momentum transfer of neutrons and Fourier time, respectively, with very high neutron energy resolution independent of the resolution of the incident neutron beam. With a pulsed neutron source, the NSE technique makes it possible to scan a wide spatiotemporal space with exceptional efficiency because both Q and τ depend on the neutron wavelength λ, which can be evaluated by the TOF technique. The Materials and Life Science Experimental Facility (MLF) at the Japan Proton Accelerator Research Complex (J-PARC) is one of the major neutron/muon experimental facilities in the world. Moreover, the Kyoto University and High Energy Accelerator Research Organization (KEK) are jointly installing two types of NSE spectrometers at BL06 at the MLF, a neutron resonance spin echo instrument (NRSE) [6] and a MIEZE [7]. The typical scientific target for the NRSE is to explore slow dynamics in soft matter, which requires a high-energy resolution with a small sample, whereas that for the MIEZE is hard matter, which requires a medium-energy resolution with a flexible sample environment, including a strong magnetic field. With the two spectrometers, it is possible to cover a wide spatiotemporal range with various sample environments. We named the spectrometers "VIN ROSE" (VIllage of Neutron ResOnance Spin Echo spectrometers). The purpose is to generate a new field of spectroscopic methods to investigate the slow dynamics of nanostructures in various materials. Both the NRSE and MIEZE make use of neutron resonance spin flippers and state-of-the-art neutron optics, which enabled us to design and install compact and multiple spectrometers in a narrow space. The performance of the supermirror guiding system for the VIN ROSE has been evaluated [8] and a 400-kHz TOF-MIEZE signal has been obtained [9]. A simple MIEZE spectrometer consists of a polarizer, a pair resonance spin flippers (RSFs), an analyzer, and a detector. There is no optical component between the sample and detector and it is easy to combine MIEZE by using the PNR and small-angle neutron scattering. The contrast of the MIEZE signal is extremely sensitive to differences between the lengths of neutron flight paths. In a continuous beam, the contrast dramatically decays by a deviation from MIEZE condition due to the misalignment from the optimal position of RSFs and detector. However, by using the TOF technique with a short-pulsed neutron beam, the misalignment effect does not change the contrast of the TOF-MIEZE signal but does change the effective frequency [10]. This means that the TOF-MIEZE signal is robust against misalignments in the instrument and it is possible to correct the misalignment with a high accuracy. However, the geometrical problem with high energy resolution measurement is still critical when the sample size and scattering angles are large [11]. On the other hand, with TOF-MIEZE reflectivity measurements, the geometrical problem is negligible even under high energy resolution measurement. There is only one spin state at the sample position and it is possible to employ polarization analysis after the sample.

3. MIEZE technique with reflectometry

The schematic of the TOF-MIEZE reflectometer and energy diagram at BL06 at the J-PARC MLF are shown in Fig. 1. In the MIEZE mode, both RSF1 and RSF2 are operated as $\pi/2$-flippers with frequencies of ω_1 and ω_2, respectively. The MIEZE signal is observed as a function of the phase difference between two energy states. The neutron intensity at the detector position is modulated by

$$\begin{aligned} I_d(t_d) &= \frac{1 + cos\phi(t_d)}{2}, \\ \phi(t_d) &= (\omega_2 - \omega_1)t_d - \frac{\omega_1}{v}L_{12} + \frac{\omega_2 - \omega_1}{v}(L_{2s} + L_{sd}) \end{aligned} \tag{1}$$

where $\phi(t_d)$ is the net phase difference between two energy states at the detector as a function of t_d. The parameters t_d and v, are time at the detector and incident neutron velocity, respectively. As shown in Fig. 1(a), L_{12} is the flight path length between RSF1 and RSF2; L_{2s} is the flight path length between RSF2 and the sample position; L_{sd} is the flight path length between the sample position and detector. In the case of the MIEZE echo condition, $\omega_1 L_{12} = (\omega_2 - \omega_1)(L_{2s} + L_{sd})$, Eq.(1) is given by $\phi(t_d) = (\omega_2 - \omega_1)t_d$.

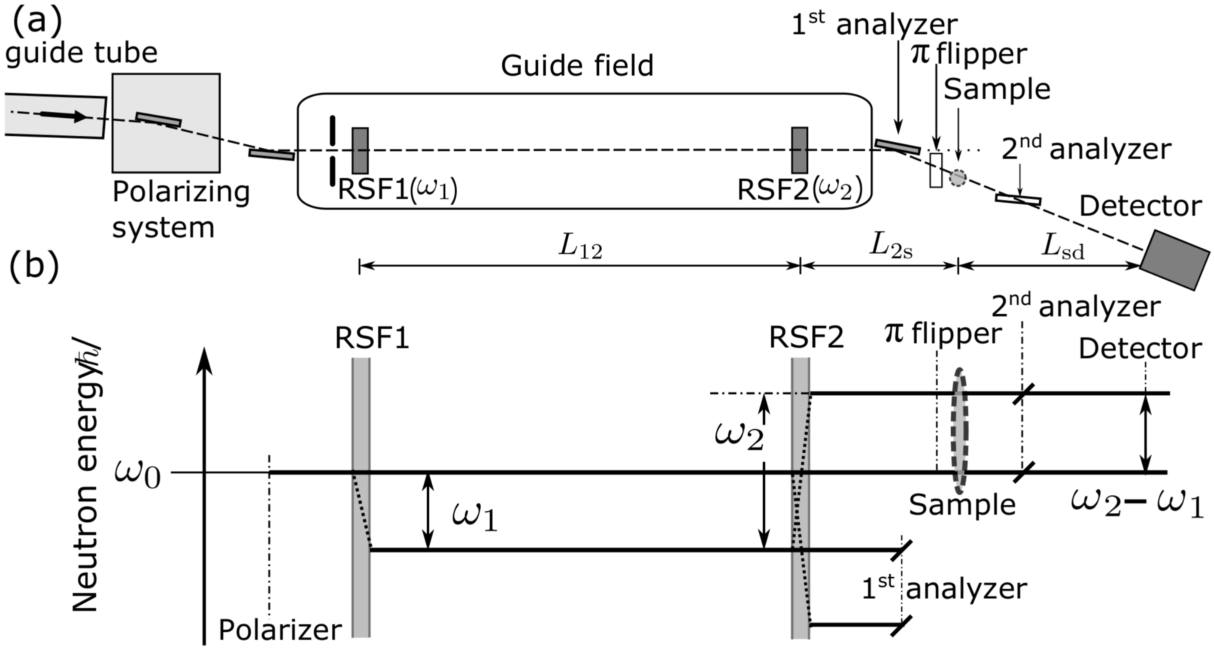

Figure 1. (a) Schematic of the experimental setup and (b) Energy diagram of the TOF-MIEZE reflectometer at BL06 at J-PARC MLF.

The energy resolution of an MIEZE spectrometer is also described by the Fourier time in the same as for NSE and NRSE spectrometers. The MIEZE Fourier time τ, is given by

$$\tau = \frac{\hbar L_{sd}}{m_n v^3}(\omega_2 - \omega_1) \tag{2}$$

where m_n is the mass of the neutron and $\hbar$ is the reduced Planck constant. Here the phase difference with the sample in the MIEZE condition is given by the following equation: $\phi(t_d) = (\omega_2 - \omega_1)(t_d + \frac{\delta L_{sd}}{v})$, where L_{12}, L_{2s}, and L_{sd} are 2.4, 0.75, and 1.2 m in the MIEZE condition, respectively. The absolute value $|\delta L_{sd}/v|$ decays the contrast of the MIEZE signal without neutron velocity change by a sample. Therefore it should be smaller than half of period of the MIEZE signal.

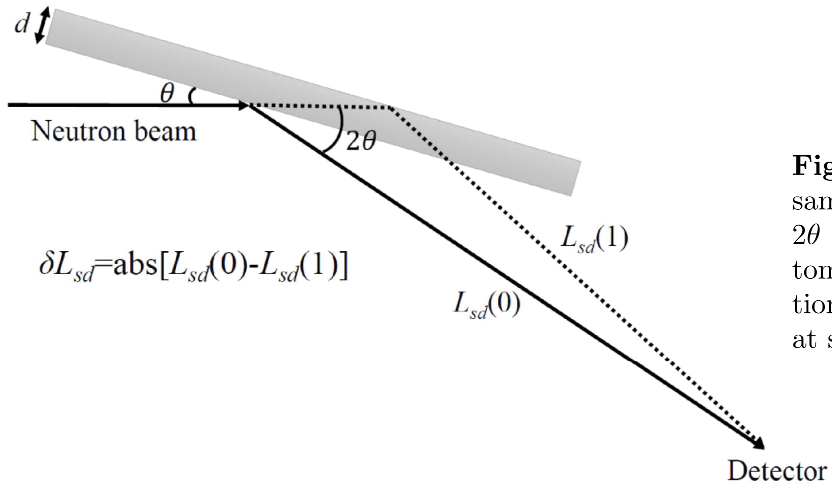

Figure 2. Schematic of the sample-detector geometry ($\theta-2\theta$ configuration) for reflectometry and a possible deviation of the path length, δL_{sd}, at same detector position.

Let's us consider a geometrical problem with MIEZE reflectometry. Here we focus on path length deviations in the reflection. A sample-detector geometry, which is called the $\theta - 2\theta$ configuration, is shown in Fig. 2. The deviation of the path length, δL_{sd}, is described by the absolute value of the difference between path lengths at the sample detector position.

$$\delta L_{sd} \quad = \quad L_{sd} - \left(\sqrt{\left(L_{sd}\cos 2\theta - \frac{d}{\cos\theta} \right)^2 + (L_{sd}\sin 2\theta)^2} + \frac{d}{\cos\theta} \right) \qquad (3)$$

With neutron reflectometry, the sample thickness and incident angle are smaller than 10 μm and 10°, respectively. In this experimental setup (L_{sd} =1.2 m) and Eq.(3), the deviation of the path length δL_{sd} is 0.61μm. Here we consider extra deviation of the path length due to the spatial resolution of detector. Which is given by $\sqrt{L_{sd}^2 + (\delta s/2)^2} - L_{sd} \sim 0.1\mu$m, where δs is the spatial resolution of detector and the deviation of the path length δL_{sd} is smaller than 1μm. The period of a 10-MHz MIEZE measurement with 20 nm wavelength neutrons is estimated to be 19.8 μm and it is 20 times larger than the deviation of the path length. Here the Fourier time given by Eq.(2) is estimated to be 613 ns. The geometrical problem with MIEZE reflectometry is negligible.

4. Experimental results and discussion

A typical TOF-MIEZE signal in which the effective frequency is 200 kHz, is shown in Fig. 3(a). The total flight path length from the neutron source to the detector is 23 m. In this experimental setup, the effective Fourier time was evaluated to be from 65 ps∼2.6 ns, at time-of-flights, from 20*to* ∼70 ms, which corresponds to the neutron wavelengths between 0.35 and

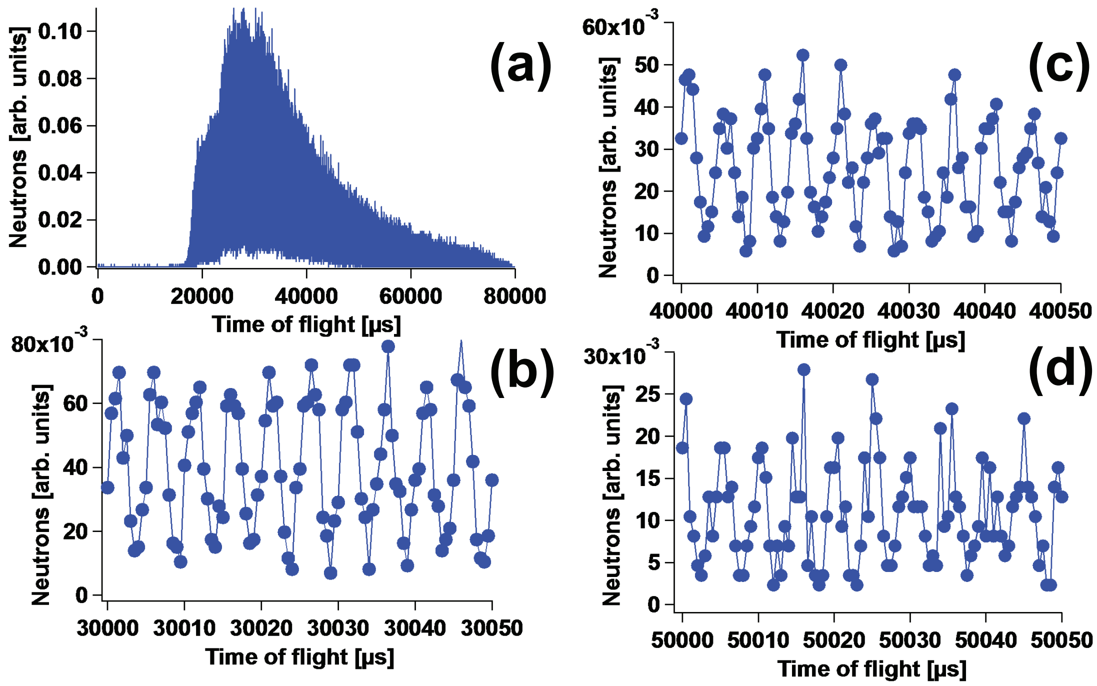

Figure 3. (a) Typical TOF-MIEZE signal with an effective frequency of 200 kHz at the BL06 at J-PARC MLF. The details of TOF-MIEZE signals in three TOF regions from (b) 30∼30.05 ms, (c) 40∼40.05 ms, and (d) 50∼50.05 ms.

1.2 nm, respectively. The frequencies of the oscillating fields of RSF1 and RSF2 were 200 and 400 kHz, respectively. Sinusoidal MIEZE signals were clearly observed with a time range of 50 μs in the whole TOF range as shown in Fig. 3(b)∼(d). By using a fast Fourier transform (FFT) over the whole TOF frame (80 ms), a clear peak in a power spectrum was observed at 200 kHz. Under the same instrumental condition, the TOF-MIEZE signal of up spin neutrons reflected by the thick iron film in which thickness 50 nm under an external field of 60 mT was observed (Fig. 4(a)). Here, the up spin and down spin are parallel and antiparallel to the magnetic guide field, respectively. The FFT analysis shown in Fig. 4(b) confirmed that the effective frequency of the MIEZE signal kept 200 kHz. Also, sinusoidal MIEZE signals were clearly observed with a time range of 50 μs even in the TOF region presenting Kiessig fringes shown in 4(c), interference originated from the film thickness. There is no evident decay in the contrast of the TOF-MIEZE signal because it is thick and the external magnetic field is sufficiently strong to magnetically saturate (4(d)).

In order to select spin flipped and non-flipped neutron intensities, a second analyzer system was installed around the sample. The analyzer system consists of a π flipper and m=4 Fe/SiGe$_3$(Si) magnetic supermirror [12]. The additional π flipper was placed between the first analyzer and sample. The second analyzer mirror, used in a transmission geometry, was placed after the sample. Here, down spin neutrons can be transmitted through the second analyzer. A magnetic guide field was extended from the first analyzer to the second analyzer in order to avoid the depolarization of the neutron spin. Under these experimental condition, ($\boldsymbol{P} \perp \boldsymbol{Q}$), the polarization vector of neutron $\boldsymbol{P}$, is perpendicular to the scattering vector $\boldsymbol{Q}$. The magnetic scattering produces spin-flip scattering. When an incoherent nuclear scattering is negligible, the intensities of magnetic (I_{Mag}) and nuclear scatterings (I_{Nucl}) are given by the following equations: $I_{Mag} = 2I_{OFF}$, $I_{Nucl} = \left(I_{Nucl} + \frac{I_{Mag}}{2}\right) - \frac{I_{Mag}}{2} = I_{ON} - I_{OFF}$

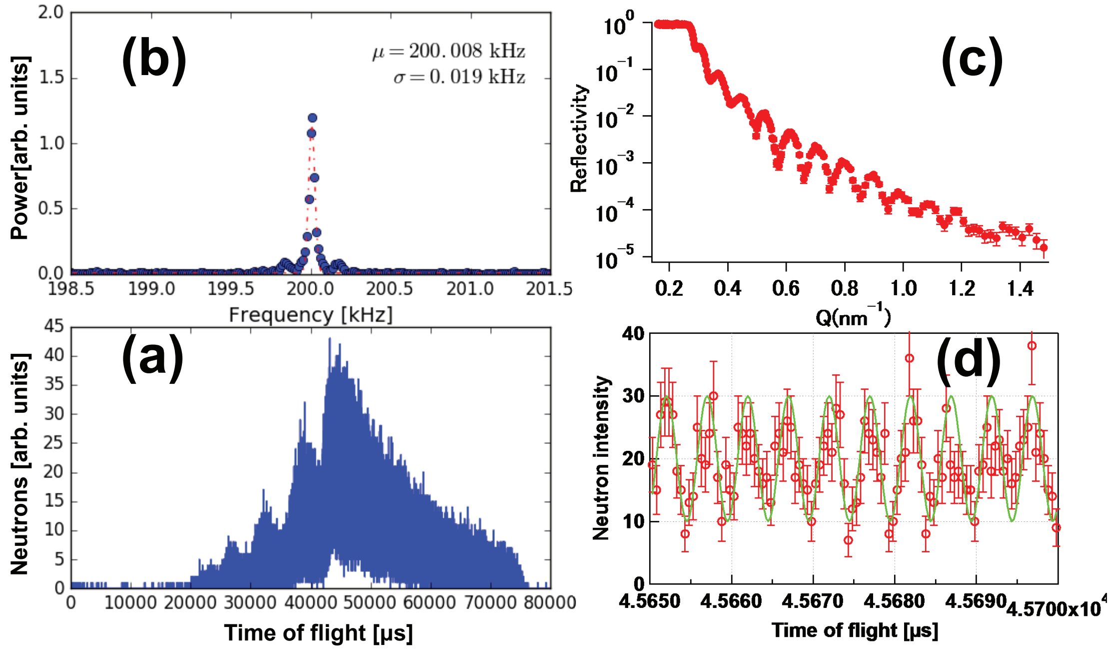

Figure 4. (a) Measured TOF-MIEZE signal reflected by the Fe thick film under an external 60-mT magnetic field. (b) The power spectrum of the TOF-MIEZE signal (c) Neutron reflectivity of up spin neutrons derived from the average of the TOF-MIEZE signals. (d) TOF-MIEZE signals from 45.65∼45.7 ms in the TOF region.

where the intensities of the non-spin flip (I_{ON}) and spin-flip scattering (I_{OFF}) are with and without π-flip, respectively. We attempted to prepare a Fe/Si multilayer in which the Fe layer is superparamagnetic at room temperature. The multilayer was fabricated using an ion-beam sputtering machine at the Institute for Integrated Radiation and Nuclear Science, Kyoto University [13]. The nominal thicknesses of Fe and Si were 0.3 and 11.7 nm, respectively. The number of bilayers was 500. The TOF-MIEZE signals of spin-flip and non-spin flip neutrons reflected at the Fe/Si multilayer surface are shown in Fig. 5(a) and Fig. 5(b). The strength of the external magnetic field at the sample position was less than 1 mT. The distinct Bragg peaks of non-spin flip neutrons and power spectrum converted from the TOF-MIEZE signal are shown in Fig. 5(a). The peak at the power spectrum corresponds well to the effective frequency of 200 kHz set by RSF1 and RSF2. In Fig. 5(b), it is also observed that the distinct Bragg peaks of spin-flip neutrons and power spectrum were converted from the TOF-MIEZE signal. Similarly, the peak at the power spectrum corresponds to the effective frequency of 200 kHz. The coincidence of these frequencies indicates that the echo condition of the MIEZE signal, sample geometry, and static magnetic field are satisfied. By accumulating individual oscillations of the TOF-MIEZE signal within a few milliseconds, it is possible to evaluate its contrast [14]. However, it is not easy to quantitatively show the contrast of the TOF-MIEZE signal because of the low neutron intensity and relatively high frequency of 200 kHz. Thus we tested a new procedure to extract contrast of TOF-MIEZE signal precisely as follows. The contrast of MIEZE signal C is defined in equation 4,

$$C = \frac{A}{B} = \frac{2|\Sigma_{k=0}^{\nu N_{div}} I_k \exp(-2i\pi \frac{k}{N_{div}})|}{\Sigma_{k=0}^{\nu N_{div}} I_k} \tag{4}$$

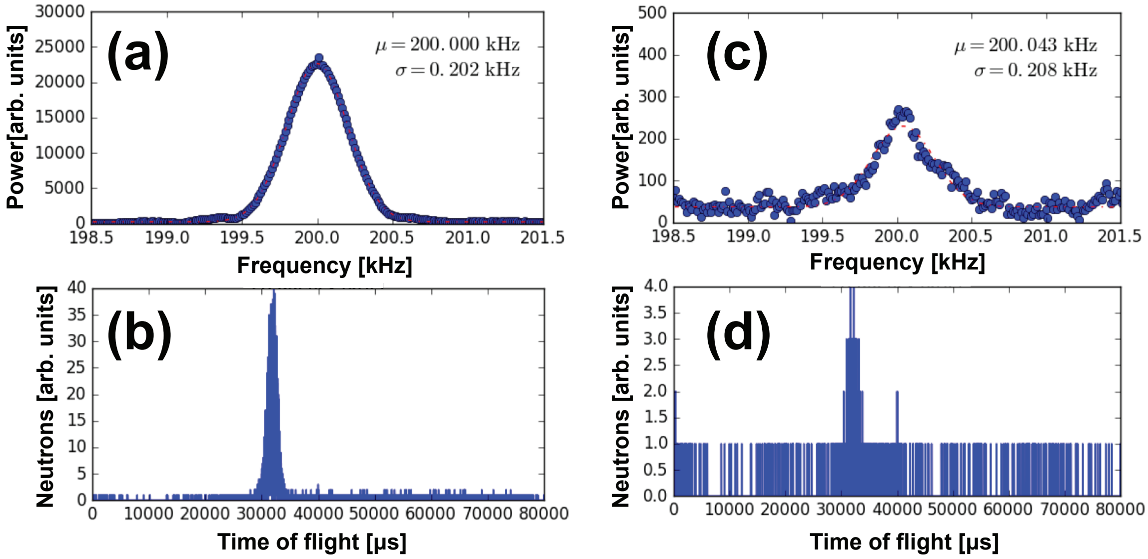

Figure 5. (a) Power spectra and (b) TOF-MIEZE signals reflected by the Fe/Si multilayer with non-spin flip (I_{ON}). (c) Power spectra and (d) TOF-MIEZE signals reflected by the Fe/Si multilayer with spin flip (I_{OFF}).

where A is twice of the amplitude of the MIEZE signal, B is the average neutron intensity, ν is the number of the MIEZE signal period, N_{div} is the division number of the MIEZE signal period and I_k is the neutron intensity at the micro region of k. $\Sigma_{k=0}^{\nu N_{div}} I_k$ represents a summation of the neutron intensity in the micro region. $|\Sigma_{k=0}^{\nu N_{div}} I_k \exp(-2i\pi \frac{k}{N_{div}})|$ represents an absolute value of a discrete Fourier transform (DFT) for the neutron intensity. The amplitude of the DFT (half of the parameter A) corresponds to the number of events which are oscillation components of the MIEZE signal. In the ideal conditions that all events are regarded as oscillation components of the MIEZE signal, the Contrast is close to 1. The parameter A and the parameter B are treated statistically from the definition. As an error handling, the error of the Contrast is based on an error propagation as follows, $\sigma_c = \sqrt{\frac{\sigma_A^2}{B^2} + \frac{A^2\sigma_B^2}{B^4}} = \sqrt{\frac{2A}{B^2} + \frac{A^2}{B^3}}$ where σ_A is the error of the parameter A, σ_B is the error of the parameter B. σ_A and σ_B represent a $\sqrt{2A}$ and a $\sqrt{B}$, respectively.

Figure 6 shows the contrast of MIEZE signal estimated as a function of time of flight, where the time bin was 0.5 μs, N_{div}=10 and ν=160 in Eq.(4). The region of interest (ROI) area in the detector is different from that in Fig. 5, and it is determined by the following condition: the number of neutrons is not zero and the 200kHz -MIEZE oscillation can be observed. In the ROI area, the instrumental background intensity owing to imperfectness of the additional pi flipper and second analyzer also makes 200kHz-MIEZE oscillation because the background intensity came from non-spin flip component. We analyzed it paying attention to the neutron statistics and show only statistically significanat data shown in Fig. 6 in the TOF time rage of 30$\sim$34 ms. We observed decay of contrast of MIEZE signal spin flipped neutrons by the Fe/Si multilayer. The neutron wavelength at time of flight 32 ms was 0.55 nm and the Fourier time was estimated to be 0.26 ns. This result shows the combination of the TOF-MIEZE technique

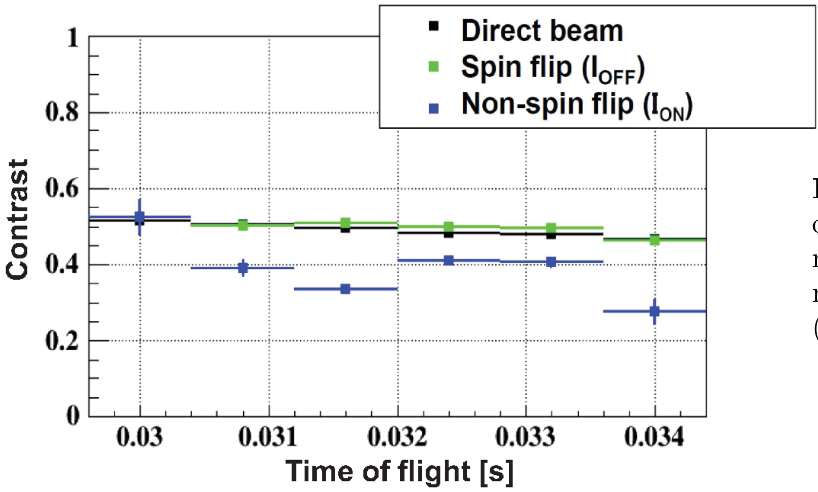

Figure 6. The contrast of MIEZE signal with direct beam, reflected by Fe/Si multilayer with non-spin flip (I_{ON}) and spin flip (I_{OFF}).

and polarized neutron reflectometry enables us to study the nanomagnetic spin dynamics in a thin film.

5. Summary

The combination of the polarized neutron reflectometry and TOF-MIEZE technique makes it possible to study the spin dynamics of the in-plane magnetization in thin films. The experimental results of TOF-MIEZE signals reflected by the Fe thin layer and Fe/Si multilayer with the second analyzer system were shown. Employing the polarized neutron reflectometry, the effective frequency of 200 kHz set by RSF1 and RSF2 is well reproduced as the peak of the power spectrum converted from the TOF-MIEZE signal. Similarly, using the second analyzer system, even under a sparse intensity, the effective frequency is also observed as the peak of the power spectrum converted from the TOF-MIEZE signal. This means that the combination of the TOF-MIEZE reflectometry and second analyzer system works similarly. Moreover, polarizing the supermirror even with extremely thin magnetic layers, makes it possible to use it as the second analyzer. We observed the decay of contrast of MIEZE signal spin flipped neutrons by the Fe/Si multilayer with Fourier time 0.26 ns.

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