International Journal on Magnetic Particle Imaging
Vol 11, No 1, Suppl 1, Article ID 2503051, 3 Pages

Proceedings Article

A novel method for magnetic nanoparticles deep optical imaging using SPAD

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Abstract

This paper proposes a novel magneto-optical imaging method that utilizes the superparamagnetic and optical properties of magnetic nanoparticles (MNPs). By measuring the rotation angle of linearly polarized light passing through MNPs under an external alternating magnetic field via the Faraday effect, we address the issue of optical signal attenuation in deep biological tissue imaging. We introduce a single-photon avalanche diode (SPAD) to detect extremely weak optical signals. Experimental results show that the harmonic signals of the Faraday rotation angle are linearly related to MNPs concentration. Under conditions with thick phantoms simulating deep tissue in the optical path, SPAD can still effectively detect polarization characteristics, verifying the feasibility of using the magneto-optical effect for high-resolution in vivo detection and imaging of deep biological tissues. This provides an exciting possibility for achieving high-resolution in vivo detection and imaging of deep biological tissues in the future.

I. Introduction

Magnetic nanoparticles (MNPs) have demonstrated significant potential in the field of biomedical imaging due to their unique magnetic and optical properties [1]. Magnetic Particle Imaging (MPI), first proposed in 2005, employs MNPs as tracers, enabling various three-dimensional imaging functions such as tumor imaging and targeted drug monitoring [2]. However, the imaging resolution of MPI is constrained by the magnetization properties and relaxation effects of MNPs, making it challenging to achieve sub-millimeter resolution [3].

In contrast, optical imaging can achieve resolution close to the diffraction limit, with the potential for further improvement through super-resolution techniques [4]. However, in conventional optical imaging, factors such as background absorption and tissue scattering often obscure the effective signal, resulting in a low signal-to-noise ratio and limited imaging depth. By employing magnetic nanoparticles (MNPs) as contrast agents and applying an external alternating magnetic field, the magnetic and optical properties of the MNPs are induced to vary periodically. Specifically, the magnetic field regulates the arrangement or chain formation of the MNPs, leading to a periodic modulation of the optical signals induced by the MNPs (e.g., transmission intensity, polarization rotation) with the magnetic field. This enables the extraction of signals that vary exclusively with the magnetic field, effectively “filtering out” the static background absorption or scattering. Consequently, optical methods for specific imaging of MNPs can overcome material-related resolution limitations while maintaining high contrast with background signals.

We propose a novel magneto-optical imaging method that leverages the superparamagnetic and magneto-optical properties of MNPs to achieve high-resolution biological imaging, validated through experiments using MNP solutions. To address signal attenuation issues encountered in deep-tissue imaging, we introduce a Single-Photon Avalanche Diode (SPAD) detector, enhancing imaging depth. Phantom experiments have verified its capability in weak signal extraction, and further studies on imaging performance are ongoing.

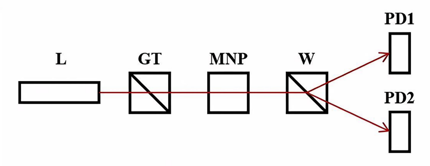

Figure 1: Schematic diagram of Faraday rotation detection device.

II. Methods and Materials

The fundamental principle of the experiment is the Faraday effect, which states that when linearly polarized light passes through a magnetized medium, the different propagation speeds (i.e., refractive indices) of the left- and right-circularly polarized components result in a phase difference between them. This phase difference subsequently leads to the rotation of the polarization plane of the transmitted light, with the rotation angle defined as the Faraday rotation angle. Essentially, the magneto-optical Faraday effect arises from the differential phase delay experienced by the two polarization components. However, this does not imply that there is an overall noticeable time delay across the entire light beam; rather, it indicates that there exists a relative phase difference within the polarization components.

In the experimental setup, uniformly distributed monochromatic light is first directed onto a sample of MNPs contained in a cuvette. The MNPs are then excited by an external sinusoidal alternating magnetic field, and a detection system collects the light signal transmitted through the MNP sample. As shown in Figure 1, L represents the helium-neon laser, GT is a Glan-Taylor prism, MNP is the sample, W is a Wollaston prism, and PD1 and PD2 are differential inputs of a balanced photodetector (PD). The axes of the Wollaston and Glan-Taylor prisms are set at an angle of $\pi/4$. To analyze the signal's frequency domain characteristics, we apply a Fast Fourier Transform (FFT) to the time-domain light intensity signal, thereby obtaining frequency-domain information and extracting harmonic signals corresponding to the magnetic field excitation frequency. This approach allows for retrieving harmonic amplitude information that reflects the MNPs concentration. In the experiments, we used diluted SHP-30 ferrite magnetic nanoparticles (Ocean NanoTech) in a water base as magneto-optical markers.

Most ferrite nanoparticles exhibit similar magnetic and optical properties, and they display analogous optical behaviors in the visible/near-infrared range. In our experiments, we selected SHP-30—a widely used Fe_3O_4 nanoparticle—by leveraging the representative characteristics common to other MNPs, thereby ensuring the broad applicability of our experimental results.

The magnetization process of MNPs is primarily influenced by two relaxation mechanisms:

- **Néel relaxation**: This refers to the reorientation of the magnetic moment within the particle without any overall translational motion of the particle itself.
- **Brownian relaxation**: This involves the overall rotation of the particle due to thermal motion, which leads to a change in the direction of its magnetic moment.

In this work, the selected MNPs have a relatively small size, which leads to the dominance of Néel relaxation. Consequently, even under the influence of an alternating magnetic field, their magnetization can rapidly follow the external field variations, resulting in a negligible phase delay.

To address the issue of significant light attenuation encountered in deep tissue imaging within biological samples, we introduced a thicker phantom to simulate such conditions and replaced the PD with a SPAD. This setup enabled the development of a weak signal harmonic extraction system for detecting extremely weak optical signals. The SPAD is used for detecting weak optical signals, and the photon pulse signals are processed through counting and FFT operations on an FPGA, ultimately yielding the harmonic signal of the light signal.The light intensity attenuation introduced by the phantom can essentially simulate the attenuation observed in biological tissues, and the attenuation factor generally follows the Lambert–Beer law. According to this law, the decay of light intensity with the optical path length can be expressed as

$$I = I_0 \exp(-\alpha L)$$

where I_0 is the incident light intensity, I is the transmitted light intensity, α is the absorption coefficient of the medium, and L is the optical path length.

III. Results and Discussion

In solutions with varying concentrations of MNPs, we successfully detected the fundamental and higher-order harmonic signals of the Faraday rotation angle. As the concentration of MNPs increased, the amplitude of each harmonic increased linearly. As previously reported by

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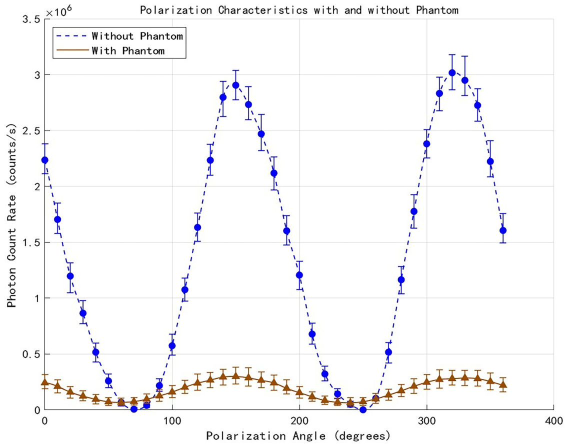

Figure 2: Experimental results show that the amplitude of the optical signal experiences significant attenuation after introducing the phantom; however, the polarization characteristics remain essentially unchanged.

our lab group [5], this finding demonstrates that the concentration of MNPs can be quantitatively measured by detecting the magneto-optical Faraday rotation angle.

When a thick phantom was introduced in the optical path to simulate deep biological tissue conditions, the signal from conventional detectors decayed rapidly, making it difficult to capture effective signals. However, by employing single-photon weak light detection technology, higher-order harmonic signals were still detectable even within a 2 cm thick phantom, with a significantly improved signal-to-noise ratio (SNR). As shown in Figure 2, despite the strong attenuation in light intensity after the introduction of the phantom, the transmitted light retained the same polarization characteristics. All amplitude values represent the first harmonic amplitudes extracted using the FFT, with the harmonic corresponding to the light signal modulated at a frequency of 100 Hz. In the presence of the phantom, although the transmitted light undergoes significant absorption at the phantom site, the overall polarization characteristics of the light intensity remain unchanged. This indicates that the method is feasible under varying light intensities.

IV. Conclusion

The proposed magnetic particle optical imaging technique has successfully achieved high-contrast detection of MNPs in vitro. By incorporating a single-photon detector, we validated the feasibility of using the magneto-optical effect to detect MNPs in deep biological tissue imaging. Future research will focus on optimizing the detection accuracy and noise performance of the single-photon detector, selecting appropriate wavelengths, and further advancing quantitative detection and imaging of MNPs in vivo.

Acknowledgments

Research funding: This work was supported by the Strategic International Cooperation Special Project of the Ministry of Science and Technology Key R&D Program: Measurement and Control Technology "Belt and Road" Joint Laboratory Construction and Joint Research on Temperature Imaging Technology (Grant No. 2022YFE0204700), with a project duration from November 2022 to October 2025.

Author's statement

Conflict of interest: Authors state no conflict of interest. Informed consent: Informed consent has been obtained from all individuals included in this study. Ethical approval: The research related to human use complies with all relevant national regulations, institutional policies and was performed in accordance with the tenets of the Helsinki Declaration, and has been approved by the authors' institutional review board or equivalent committee.

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