LETTER

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Transient magneto-optic effects in ferrofluid-filled microstructured fibers in pulsed magnetic field

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Abstract – Transient magneto-optic effects in ferrofluid-filled microstructured optical fibers are considered. Magneto-optic responses of two types, *i.e.*, an even and an odd one, were observed in the longitudinal geometry of an applied pulsed magnetic field for the kerosene-based Fe_3O_4 ferrofluid with ∼8 nm nanoparticles. For the first time a submicrosecond response time limited by the rise time of the applied field pulse (∼0.35 μs) was demonstrated for the odd magneto-optic effect in an all-fiber system, and responses of the even and odd magneto-optic effects were separated. A strong influence of the pulse width on the relaxation time of the even response is attributed to the formation of particle aggregates.

Magnetic fluids (MF), or ferrofluids, which are suspensions of magnetic nanoparticles belonging to the class of the so-called "soft matter" are interesting objects for research and attractive for different applications [1–3]. Considerable attention has been given to the magneto-optic properties of these materials [4–6]. The behavior of such fluids in an external magnetic field was discussed in [7], where processes of field-induced aggregation of pre-existing large aggregates and orientation of few-particle pre-existing aggregates were considered. The formation of long (in comparison with light wavelength) chain-like aggregates was studied using optical microscopy [8]. Dynamic measurements of magneto-optic effects in more stable magnetic fluids [9] have shown that the transverse effect arises due to the Brownian orientation of particles having shape anisotropy in a magnetic field. The longitudinal effect results from either the rotation of a particle or orientation of magnetic moment in superparamagnetic nanoparticles via the Néel relaxation. Potentially, a very fast response of the longitudinal effect can be achieved, and it was proposed that it be used for optical modulation up to significant frequencies (> 1 MHz) [4].

The magneto-optic effect in magnetic fluids can be enhanced by light waveguiding in an optical fiber. The light-MF interaction in an optical fiber can be achieved by two basic methods, *i.e.*, by inserting a conventional fiber with a removed cladding or tapered fiber directly into a magnetic fluid [10–12] (in its turn, the fiber can be placed into a capillary of a larger diameter) or by infiltrating a microstructured fiber with a magnetic fluid [13–15]. These layouts are convenient for MF insulation/sealing and for optical coupling with an external optical fiber or an integrated optics circuit. The all-fiber optical configuration is promising for the development of new photonic devices used to control transmitted-light parameters, such as sensors, modulators, switches, optical logic elements, etc. The fast operation of MF-based optoelectronic systems is highly desirable and all-fiber configurations can be expected to contribute to short response times. However, the experimentally demonstrated characteristic switching times of MF-based fiber-optic devices are typically not shorter than 10^{-3} s [16,17]. For example,

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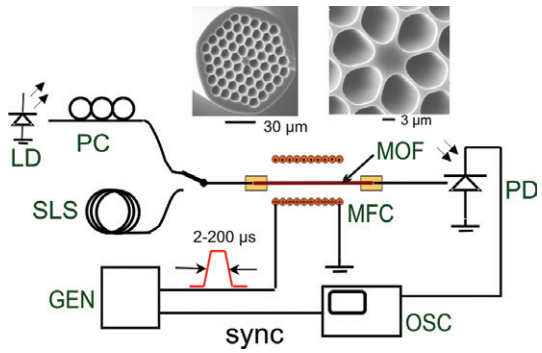

Fig. 1: (Colour on-line) Experimental configuration: LD is the cw semiconductor laser diode, SLS the erbium-doped-fiber–based superluminescent source, PC the polarization controller, MOF the microstructured fiber, MFC the magnetic-field coil, PD the photodiode, GEN the signal generator, OSC the oscilloscope. The inset shows the fiber cross-sections.

the magneto-optic logic element with a water solution of ∼40 nm Fe_3O_4 particles reported in [16] had a characteristic response time of about 10 ms, the additional waiting time being on a similar scale. A minimum switching time of about 0.3 ms was reported for the water-based ferrofluid with ∼10 nm Fe_3O_4 particles in [17].

Our group has recently reported the measurements of the dynamic magneto-optic response from a microstructured fiber with a silica core infiltrated with a kerosene-based MF [18] which agree with the results of [9] and show that the magneto-optic response can be much faster. This paper deals with the experimental investigations of the transient response of a similar microstructured fiber-based MF cell under a pulsed excitation in a longitudinal geometry of the magneto-optic effect.

The data presented below were obtained for a 3 cm long segment of a microstructered optical fiber with a 7 μm silica core (see the inset to fig. 1). The fiber cladding hollow channels were filled with a MF by means of a capillary force. To provide the long-term stability of the cell and prevent drying of the MF, the channels were sealed by a special epoxy with the refractive index lower than that of silica. Then single-mode SMF-28 fibers were butt-coupled to the cell input and output with a UV curable adhesive. It provided stable launching conditions and easy integration into the fiber system, which is very useful for possible applications.

The main criterion for the magnetic-fluid choice was its applicability to infiltration of a microstructured fiber. Fe_3O_4 nanoparticles with a median diameter of 8 nm and scaling parameter of 0.17 of the lognormal distribution were synthesized from the solution of $FeCl_3$ and $FeSO_4$ by precipitation with ammonia water and subsequent filtering, washing, high-temperature drying and magnetic separation. Oleic acid was added as a surfactant to prevent their permanent aggregation. We selected kerosene as a solvent because of the absence of absorption bands in the near infrared due to a low fundamental vibration energy. The additional advantage of kerosene is a low viscosity. A volume concentration of ∼0.13% gave an average MF refractive index of ∼1.398 in the wavelength range 1520–1540 nm. The MF-filled fiber fundamental mode diameter for this refractive index was estimated to be 6.8 μm. This implies an approximately 4% overlap of the mode intensity profile with the filling magnetic substance [19]. This concentration provides a trade-off between the strength of interaction and optical losses. The total transmittance of the all-fiber cell prepared in this way was ∼ −11 dB, which was mostly caused by coupling losses due to mode mismatch with input and output SMF-28 fibers.

The experimental setup (fig. 1) was similar to that described in [18]. Both the even and odd magneto-optic responses could be studied in this configuration. Light sources of two types were used, *i.e.*, a DFB semiconductor laser with a polarized output at 1532 nm and a superluminiscent erbium-fiber–based source with an unpolarized output in the 1520–1540 nm range. When the polarized source was used, the light polarization state at the cell input was set by a fiber-optic polarization controller. No polarizers were installed in front of a photodiode. This means that we observed the effects of the magnetic-field–induced circular dichroism (due to the Faraday effect) and polarization-independent transmitted power modulation (due to the Cotton-Mouton effect in the longitudinal configuration) simultaneously [18]. The external magnetic field was produced by the coil which was fed by a homemade oscillator generating driving rectangular current pulses. The pulse width could be changed from 2 to 200 μs. The minimum current pulse rise/decay time (10%–90%) of about 0.35 μs was determined by the output oscillator resistance. The solenoid was 5 mm in diameter and ∼3 cm in length. The latter was approximately equal to that of the MF-filled fiber cell inserted into the coil along its axis. Such a positioning provided a high homogeneity of the field H applied to the cell. For the maximum current amplitude of 17 A, H was estimated to be 2 kOe. All the measurements were performed at room temperature.

A typical magneto-optic response of the relaxation type to a 65 μs long rectangular magnetic-field pulse is shown in fig. 2(a) for the unpolarized incident light. This response proved to be of an even type, *i.e.*, it remained unchanged when the magnetic-field pulse was reversed.

The response shape proved to be essentially different if the incident probe light was polarized circularly. In this case both the even (Cotton-Mouton) and odd (Faraday) magneto-optic effects were observed. Two typical profiles obtained for 2 μs long magnetic-field pulses of the same amplitude but of opposite polarities are presented in fig. 2(b) as curves 1 and 2. The inversion of the circular light polarization changed the response shape to that observed with the opposite field polarity. By summing up and subtracting these two response profiles we could extract the even and odd components of the magneto-optic

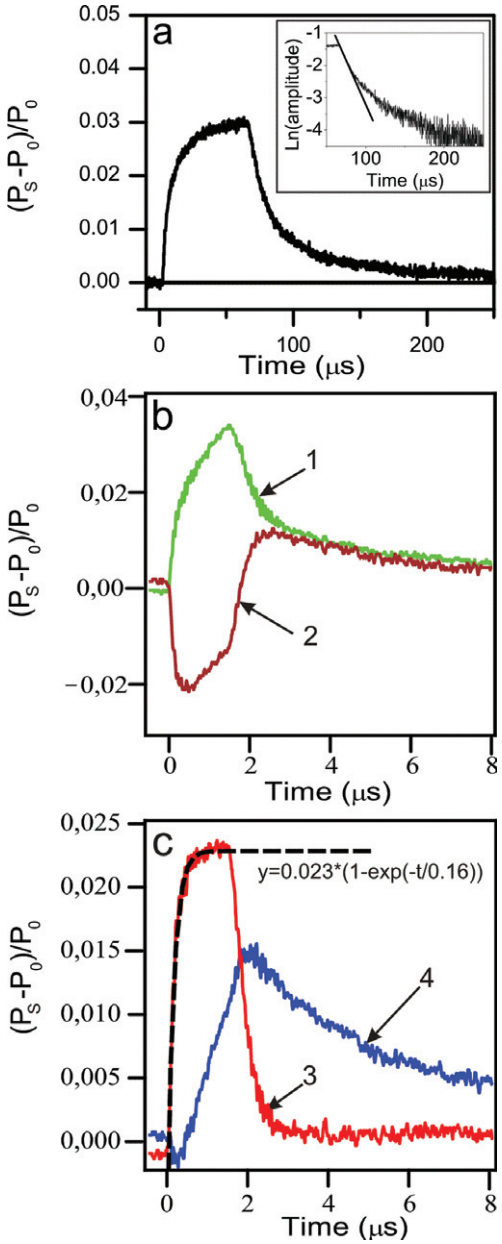

Fig. 2: (Colour on-line) Typical oscilloscope traces of the magneto-optic response: (a) for the unpolarized probe light (65 μs magnetic-field pulse), the inset shows its trailing front in the lin-log scale; (b) for the circularly polarized probe light (2 μs pulse) and for positive (curve 1) and negative (curve 2) magnetic-field directions; and (c) the odd (curve 3) and even (curve 4) components for the circularly polarized probe light and positive field (2 μs pulse), the exponential approximation of the leading front of the odd component is shown. P_0 is the initial light power, and P_S is the light power in the applied field.

response presented in fig. 2(c). Note that the even component of this response is very similar to that observed in the experiment with the unpolarized probe light.

The dependences of the steady-state values of both the odd and even magneto-optic responses (the values that are reached when transition processes are over) on magnetic

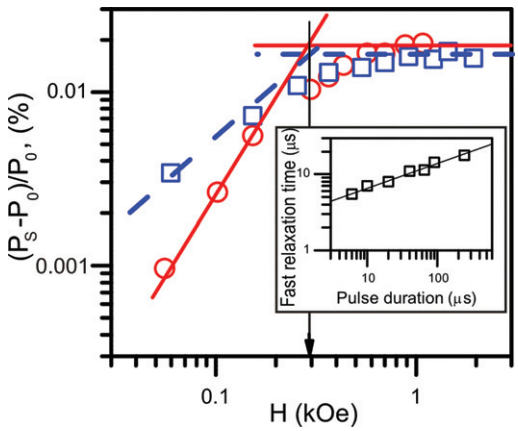

Fig. 3: (Colour on-line) Amplitude of magneto-optic response as a function of magnetic-field. The red circles show the even polarization-independent response for 200 μs magnetic-field pulses. The blue squares show the odd response under circularly polarized light illumination (multiplied by 3, for 2 μs pulses). The inset shows fast relaxation time of the trailing edge as a function of pulse duration ($H = 450$ Oe). P_0 is the initial light power, and P_S is the light power in the applied field.

field (fig. 3) exhibit saturation at $H \approx 300$ Oe, which correlates with the anisotropy field of Fe_3O_4 nanoparticles known from the literature [20].

Let us start our discussion with the odd magneto-optic response, *i.e.*, the magnetic-field–induced circular dichroism. We attribute it to rather a fast Faraday effect associated with magnetization rotation, which, at least at the first moment, occurs inside every nanoparticle with the characteristic time as short as 10^{-8} s [21]. The experimentally observed minimum duration of the odd response fronts (the characteristic time constant was about 0.16 μs, see fig. 2(c)) was limited by the pulse current generator. It can be supposed that in the case of ultrashort field pulses the even response will not be formed because of a rather long characteristic rise time, however the odd response will be preserved. This fact is important for development of MF-based fast light modulators.

Both the leading and trailing fronts of the even response (fig. 2(a)) consisted basically of the dominating "fast" (with the microsecond-scale characteristic relaxation times) components and a smaller contribution of some slower processes (see the inset to fig. 2(a)). The "fast" characteristic time of the leading front was approximately constant ($\tau \approx 4.7\,\mu$s). The even response with similar characteristic times is typically attributed to the Cotton-Mouton effect associated with rotation of individual nanoparticles [9,22]. In our earlier experiments the corresponding linear dichroism was observed as a magnetic-field–induced change in the optical absorption which was polarization-independent in the longitudinal configuration [18].

The "hydrodynamic" diameter d_h of a particle is estimated from the characteristic time of the even

magneto-optic effect by using the classical relation [22]

$$d_h = \sqrt[3]{\frac{6\tau k_{\mathrm{B}} T}{\pi\eta}}, \tag{1}$$

where η is the liquid viscosity and $k_B T$ is the thermal energy. For the average viscosity of kerosene ($0.00164\,\mathrm{Ns/m^2}$) d_h proved to be $\sim$40 nm, which is approximately five times larger than the average particle size as extracted with lognormal distribution fit of the electron microscope data $d \approx 8.2$ nm [23]. Such a discrepancy is often found in the literature [7,22]. In [22] it is explained by greater influence of large-diameter particles and particle aggregates with respect to small ones in low magnetic fields. The characteristic "fast" relaxation time of the trailing front grew from the minimum time $\tau \approx 4.7\,\mu$s with increasing applied magnetic-pulse duration (see the inset to fig. 3). This effect can be attributed to formation of more inertial clusters (chains) of magnetized particles [24]. This hypothesis is also supported by the experiments with application of a relatively strong (up to 4 kOe) transverse dc magnetic field. Application of such a field resulted in an approximately twofold reduction in the even response amplitude and also in a significant decrease in the relaxation time of the response trailing front. Probably, the strong transverse field gave rise to the formation of particle chains along its direction and prevented the formation of orthogonally oriented chains by the pulsed longitudinal magnetic field.

A more stable MF can be used to prevent aggregate formation under a magnetic field [9]. A good way to stabilize a solution of magnetic nanoparticles is to embed them in transparent matrices [25]. However, these stable magnetic substances should also be suitable for fiber infiltration and have a rather narrow range of refractive indices to provide light guiding. Another way to obtain a better stabilized magnetic fluid is to use finer particles [26]. The additional advantage of such particles is a partial suppression of the even magneto-optic effect [10]. However, the applicability of such magnetic fluids to the filling microstructured fibers depends on many factors, such as types of solvent and stabilizing agent, optical loss of the fluid, refractive index, etc. which should be additionally studied. Thus, the choice of a specialized type of magnetic substance for the fiber optic implementation is a subject of additional investigations.

To summarize, detailed experimental investigations of transient magneto-optic effects in a MF-filled microstructured fiber with a silica core were carried out for a longitudinal pulsed magnetic-field excitation. Similarly with a volume configuration [9] the behavior of the magneto-optic response was explained by the contribution of two magnetic processes in the system of nanoparticles, *i.e.*, the Faraday magnetization of individual particles and the Cotton-Mouton effect responsible for their cooperative behavior. It was found that the even magneto-optic effect (transmitted power modulation) was characterized by a constant pulse rise time and an appreciable growth in the pulse decay time with increasing magnetic-pulse duration. This, together with the suppression of this effect by the application of a strong transverse magnetic field, led us to the conclusion that a significant contribution into the even effect came from the formation of magnetized particle aggregates. The odd magneto-optic response was attributed to a fast Faraday effect associated with the material magnetization. Thus, the submicrosecond magneto-optic modulation in an MF-based device was experimentally demonstrated for the first time. It is at least two orders of magnitude faster than the literature data available at present. The results can be applicable to a wide variety of MF-based structures, and, in particular, to the optofluidic technology.

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