Spectral Properties of Magneto-plasmonic Nanocomposite. Vertical Shift of Magneto-Optical Hysteresis Loop

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Abstract. In present work the results about study of spectral features of the magneto-optical hysteresis loop (Faraday rotation) vertical shifting effect in the magneto-plasmonic nanocomposite Bi:YIG/Au(NP)/GGG are shown. It is shown that in the spectral range near localized plasmon-polariton resonance (LPPR) the magneto-optical hysteresis loop has a vertical shift (Faraday “delta”). The loop shifting at the left and right side of the LPPR have a different sign and in the resonance the loop shifting become null. It is shown that the spectral locations of Faraday “delta” maxima are due to the width of the LPPR resonant line.

1. Introduction
Magneto-optical sensing systems and structures are widely used in control devices of fiber-optic communication lines, in manufacturing of sensitive magnetic field sensors for magnetocardiography and low-field magnetic tomography, in visualizing of magnetic fields for defects detection by eddy current, etc. [1]. One way to enhance the sensitivity of such systems and structures is enhancing the specific magnitude of magneto-optical effects (in particular the Faraday effect). There is a method to enhancing the Faraday effect by injecting the plasmon subsystem into the magneto-optical dielectric medium, which becoming to significantly enhancement of the Faraday rotation on the range of plasmon resonance [2]. A similar effect was also observed by the authors of this article but at sample remagnetization not only increasing of the magneto-optical hysteresis loop (MOHL) height (enhancement of the effect) was detected, but also an effect of the vertical shift of the loop (Figure 1), which named Faraday “delta”.

Figure 1. The magneto-optical hysteresis loop (MOHL) in pure bismuth-substituted ferrite-garnet film Bi:YIG/GGG (without plasmonic Au nanoparticles) and magneto-plasmonic nanocomposite Bi:YIG/Au(NP)/GGG (with plasmonic Au nanoparticles): θ_{FR} – angle of Faraday rotation at film magnetic saturation (half-height of the magneto-optical hysteresis loop); Δθ – “Faraday delta”, additive vertical shifting of the MOHL.
2. Experimental technique

The magneto-plasmonic nanocomposite Bi:YIG/Au(NP)/GGG (Figure 2a) was synthesized by method of vacuum deposition with following heat treatment. It’s a layer of self-assembled gold nanoparticles Au(NP) which formed on a gallium-gadolinium garnet (GGG) substrate and covered with a continuous layer of bismuth-substituted ferrite-garnet Bi:YIG. During materials deposition an original technique for obtaining films with a thickness gradient along the sample surface was used [3]. This technique allows to carry out studies of films with different thickness at one sample which was obtained in one technological cycle.

In this experiment we synthesized a sample with gradient of Au film effective thickness \( h_{\text{eff}}(\text{Au}) \). This sample was investigated in three areas where \( h_{\text{eff}}(\text{Au}) \) is 1.6 nm, 1.3 nm and 1 nm, respectively. Layer of bismuth-substituted ferrite-garnet Bi:YIG has constant thickness 100 nm for all sample area. Bi:YIG has Bi\(_{1.5}\)Gd\(_{1.5}\)Fe\(_{4.5}\)Al\(_{0.5}\)O\(_{12}\) stoichiometric composition.

After Au film deposition, it has been treated by annealing on air at 950\(^\circ\)C during 10 min for formation of self-assembled gold nanoparticles Au(NP)/GGG. SEM-image on figure 2b demonstrate the surface morphology of granulated Au film. As can see, the form of self-assembled gold nanoparticles is ellipsoidal.

\[ \theta_{\text{TR}} = \frac{\theta_{\text{TR}}(H^+) - \theta_{\text{TR}}(H^-)}{2}, \]

Accordingly, the shifting of the magneto-optical hysteresis loop (Faraday “delta”) was determined as the value of the midline between the spectra in the \( H^+ \) and \( H^- \) field:

\[ \Delta \theta = \frac{\theta_{\text{TR}}(H^+) + \theta_{\text{TR}}(H^-)}{2} \]
Figure 3. Scheme of spectral photometer-magneto-polarimeter for investigation of magneto-optical Faraday effect in magneto-plasmonic nanocomposite Bi:YIG/Au(NP)/GGG.

3. Results and discussion
The investigation results of optical properties of magneto-plasmonic nanocomposite and his components are shown on figure 4. So, on figure 4a the transmittance spectra of Au/GGG film after deposition (before treatment) are presents. It’s spectra for sample's areas with different effective thickness (1.6 nm – green line, 1.3 nm – red line, 1 nm – blue line).

Figure 4. Transmittance spectra in different areas of gradient of Au film effective thickness (1.6 nm – green line, 1.3 nm – red line, 1 nm – blue line): (a) The film Au/GGG after deposition; (b) The nanoisland film Au(NP)/GGG after annealing at 950°C during 10 min; (c) The magneto-plasmonic nanocomposite Bi:YIG/Au(NP)/GGG. Also, the specter of bismuth-substituted ferrite-garnet film Bi:YIG/GGG without plasmonic subsystem is shown (violet line).
The transmittance spectra of granulated nanoisland film Au\textsubscript{NP}/GGG after annealing on air at 950°C during 10 min are shown on figure 4b. As can see, the local minima are presents in spectra. These minima are associated with energy absorption for resonant excitation of localized plasmon-polaritons in self-assembled gold nanoparticles. The resonant wavelength $\lambda_{LPPR}$ of localized plasmon-polariton resonance (LPPR) is 565 nm for all areas of Au effective thickness gradient.

On figure 4c the transmittance spectra of magneto-plasmonic nanocomposite Bi:YIG/Au\textsubscript{NP}/GGG are shown. Also, the specter of bismuth-substituted ferrite-garnet film Bi:YIG/GGG without plasmonic subsystem is shown for comparation (violet line). As can see, after deposition of 100 nm bismuth-substituted ferrite-garnet film on surface of self-assembled Au nanoparticles, the resonant wavelength $\lambda_{LPPR}$ is shifted to the long-wave spectral range. The new resonant wavelength $\lambda_{LPPR}$ = 685 nm. The spectral dip in short-wave range (less than 550 nm) is due to absorption of ferrite-garnet film.

The spectral features of magneto-optical properties of magneto-plasmonic nanocomposite Bi:YIG/Au\textsubscript{NP}/GGG are shown on figure 5.

For comparation, on figure 5a the magneto-optical properties of pure ferrite-garnet film Bi:YIG/GGG without plasmonic subsystem are presents. The spectra of magneto-optical total rotation of the transmitted radiation polarization angle $\theta(\lambda)$ are shown there. They were obtained during sample magnetization in the $H^+$ field, directed along the radiation wave vector $k$, and in the $H^-$ field, directed against the wave vector. The Faraday rotation specter, determined by equation (1), coinside with specter of total rotation in saturation field $H^+$, because the spectra in fields $H^+$ and $H^-$ are symmetrical relatively the $\lambda$-axis. In this case, the Faraday “delta” $\Delta\theta$ equal to null in all spectral range.

![Figure 5](image-url)

**Figure 5.** Spectra of magneto-optical properties of ferrite-garnet film Bi:YIG/GGG without plasmonic subsystem (a) and nanocomposite Bi:YIG/Au\textsubscript{NP}/GGG in different area of gradient of Au film effective thickness (1.6 nm – green line, 1.3 nm – red line, 1 nm – blue line): (b) The angle of total rotation of polarization plane (solid lines – in field $H^+$, dotted lines – in field $H^-$); (c) The angle of “true” Faraday rotation; (d) The “Faraday delta” and the derivative of transmittance (black line).
On figure 5b the spectra of magneto-optical total rotation in fields $H^+$ (solid lines) and $H^-$ (dotted lines) are shown for magneto-plasmonic nanocomposite Bi:YIG/Au$_{NP}$/GGG. In the range of LPPR the asymmetric of spectra is observed. This asymmetric is due to vertical shifting $\Delta \theta$ of magneto-optical hysteresis loop.

The Faraday rotation spectra $\theta_{FR}(\lambda)$ of nanocomposite Bi:YIG/Au$_{NP}$/GGG are shown on figure 5c. Those spectra are determined by equation (1). As can see, the enhancement of Faraday rotation effect is observed on $\lambda_{LPPR}$ (magenta dotted line). For example, at the area of the sample with Au effective thickness 1.3 nm (red line) the angle of Faraday rotation enhanced almost 3.5 times on $\lambda_{LPPR} = 685$ nm. ($\theta_{FR} = -0.225$ deg for Bi:YIG/Au$_{NP}$/GGG and $\theta_{FR} = -0.065$ deg for Bi:YIG/GGG).

On figure 5d the spectra of Faraday “delta” $\Delta \theta(\lambda)$ are presents. They were obtained by equation (2) as the average line between spectra of total rotation at fields $H^+$ and $H^-$. As can see, the specter of Faraday “delta” (vertical shift of magneto-optical hysteresis loop) has two extremums: the positive maximum at the “left” side of $\lambda_{LPPR}$ (650 nm, brown dotted line) and the negative minimum at the “right” side of $\lambda_{LPPR}$ (730 nm, orange dotted line). At the wavelength of plasmonic resonance the Faraday “delta” equal to null $\Delta \theta(\lambda_{LPPR}) = 0$ (685 nm, magenta dotted line).

For comparison the derivation of transmittance specter $dT/d\lambda$ also shown on figure 5d (black solid line). As can see, both maximum and minimum of Faraday “delta” are located symmetrically to extremums of $dT/d\lambda$ specter relatively the $\lambda$-axis. So, the spectral location of maximum and minimum of Faraday “delta” is determined by LPPR resonant line width.

This effect of MOHL shifting (Faraday “delta”) is associated in [4, 5] with localized plasmonic resonance (SPR-induced rotation) for s- and p-polarized waves in anisotropic system of gold nanoparticles. But authors noted that the anisotropic don’t clearly observing on SEM-images.

We think that this effect of MOHL vertical shifting is due to the constant additive to the angle of the polarization plane rotation. This additive doesn't depend on the magnitude and sign of the external magnetic field, and its magnitude is determined by the spectral position relatively to plasmonic resonance in self-assembled gold nanoparticles. The sign changes of the Faraday “delta” and the location of its maxima, which due to the width of the LPPR resonance line, can be explained as follows.

As it was shown in [3], the size distribution of self-assembled gold nanoparticles Au$_{NP}$/GGG has a quasiisymmetric form and described well by the Gaussian (Figure 6a). At resonant frequency $\nu_{LPPR}$ (absorption maximum) the resonance conditions are satisfied for nanoparticles having the most probable size (distribution maximum). For particles that are smaller than the “resonant” (red region), the self-frequency exceed the frequency of the exciting field $\nu_{LPPR}$ (“advancing” frequency), at this case the particle polarizability is always directed against the field. For particles that are larger than the “resonant” (green region), the self-frequency will be lower than the frequency of the exciting field $\nu_{LPPR}$ (“lagging” frequency), so the particles will be polarized along the field [6].

![Figure 6](image)

**Figure 6.** The change of resonant conditions for nanoparticles with different size (for explanation of Faraday “delta” features).

It is known, that near the LPPR condition, a resonating dipole (in our case it's a nanoparticle) induce a strong electromagnetic field around itself [7, 8]. This field is localized around the resonator and
penetrated into the surrounding dielectric medium to distance of about 100 nm. It's obvious that the particles whose having “advancing” and “lagging” self-frequency (relative to the frequency of the exciting field) will induce resonant fields with the opposite direction.

Let's consider the state of the resonating system when the frequency of the exciting field is \( v_{\text{LPPR}} \) (Figure 6a). At this case the number of particles with an “advancing” self-frequency is equal to the number of particles with a “lagging” self-frequency (the red and green shaded areas are equal). Thus, the resonant fields of the resonating nanoparticles will be mutually compensated and their affecting on the ferrite-garnet layer will be minimal. It explains the null value of the Faraday “delta” in the LPPR condition.

When the frequency of the exciting field is less then \( v_{\text{LPPR}} \), the strict resonant conditions are satisfied for larger particles (Figure 6b). In this case, the number of particles with an “advancing” self-frequency (red shaded area) significantly exceeds the number of particles with a “lagging” self-frequency (green shaded area). Thus, the uncompensated resonant field appear near the particles which polarized against the exciting field, and a negative MOHL shifting is observed.

When the frequency of the exciting field is more then \( v_{\text{LPPR}} \), the strict resonant conditions are satisfied for smaller particles (Figure 6c). Wherein the number of particles with an “advancing” self-frequency (red shaded area) significantly less than the number of particles with a “lagging” self-frequency (green shaded area). In this case, the uncompensated resonant field appear near the particles which polarized along the exciting field, and a positive MOHL shifting is observed.

Based on the above, the relations between the position of the Faraday “delta” maxima and the width of the LPPR resonant line becomes obvious. The maximum MOHL shifting is observed at the condition that the maximum number of nanoparticles is involved in the resonance and the number of particles with an “advancing” self-frequency are maximally exceeds the number of particles with a “lagging” self-frequency (or vice versa).

4. Conclusions
The existence of the plasmon subsystem in the magneto-plasmonic nanocomposite Bi:YIG/Au\(\text{NP}\)/GGG leads to enhancement of the Faraday effect at the LPPR frequency in 3.5 times collation with a pure ferrite-garnet film Bi:YIG/GGG. The effect of vertical shifting of the magneto-optical hysteresis loop \( \Delta \theta \) in magneto-plasmonic nanocomposite Bi:YIG/Au\(\text{NP}\)/GGG was experimentally detected. This effect was observed near the condition of plasmon resonance in the Au\(\text{NP}\) plasmonic subsystem, at that the loop shifting change the sign on the left and right side from resonance, and in the resonance \( \Delta \theta = 0 \). The spectral locations of Faraday “delta” maxima (positive and negative) are due to the width of the LPPR resonance line.

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