Original Research Article
MODIFICATION OF MAGNETIC CHARACTERISTICS OF POLYCRYSTALLINE NIFE FILMS AT THE IRRADIATION LASER PULSES AND FORMATION OF REGULAR STRUCTURE OF MAGNETIC NANOISLANDS
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Abstract.
This paper presents the results of experimental measurements of magnetic properties of thin NiFe films after irradiation with nanosecond laser pulses. The results showed that there is an optimal energy in the laser pulse, at which the magnetic film characteristics vary the most. For the film of 500 nm thick at magnetic susceptibility $\mu$ for a Nd:YAG laser at $\lambda=1064$ nm increased almost 3 times, and the coercive force $H_c$ decreased about two. For the excimer laser at $\lambda=248$ nm, we have received an increase in the magnetic susceptibility $\mu$ almost ten times while a decrease in the coercive force $H_c$ was 5-6 times. The results suggest the importance of non-thermal effects of laser radiation in the process of changing the structure and magnetic properties of magnetic films. All this shows that with the help of laser radiation it is possible to form a regular structure of nanoislands in thin magnetic nanocrystalline films.

Keywords: laser radiation, magnetic films, change in the magnetic characteristics. magnetic susceptibility, coercive force.

1 Introduction
Laser materials processing technology has a number of significant advantages, such as high speed and local heating control flexibility of the heating mode and the ability to form a large area of complex topology of the irradiated region with a very small amount of radiation points. In most cases, thermal processing is used to laser radiation. However, there are other mechanisms of action of laser radiation on the substance that can be used in practice. Stimulated laser oxidation processes of recrystallization and diffusion play an important role when irradiating thin films. These processes must not only be taken into account in the studies, but also can be used in practice. In our opinion, a possibility of using laser processing technology in thin magnetic films has good prospects. Magnetic thin films are the basic material for spintronics elements. A special study focuses on the magnetic films with a regular distribution nanoislands. Commensurability of such nanoislands atomic scale materials provides strengthening of magnetic characteristics due to the exchange interaction. Regular nanoislands distribution makes it possible to create high-density information recording media on their basis. The basic method of forming nanoislands is electron-beam lithography. However, the production of regular nanostructures using this method is complex and expensive, which increases with the size of nanometer single element and a large area of treatment.

The results of our previous studies [1] have shown that irradiation of magnetic films by laser nanosecond pulses can change their magnetic properties. However, we were not sure what physical processes cause these changes, and how they can be used in practice.

Therefore, we conducted additional experimental studies of the interaction of nanosecond laser pulses with polycrystalline NiFe films, which have enabled us to determine the mechanism of the effect of laser radiation on the roughness of surface, on the structure and magnetic properties of magnetic films. This paper presents the results of experimental measurements of magnetic properties of thin NiFe films after irradiation with nanosecond laser pulses. The results suggest the importance of non-thermal effects of laser radiation in the process of changing the structure and
magnetic properties of magnetic films. All this shows that with the help of laser radiation it is possible to form a regular structure of nanoslands in thin magnetic nanocrystalline films. The use of this laser technology makes it possible to obtain the structure of magnetic nanoslands in the large area and perform processing quality control directly at the manufacturing stage of the given structure. This laser technology can be successfully used in spintronics and in the production of magnetic carrier.

2 Experimental technique and results

We investigated nanocrystalline Ni$_{81}$Fe$_{19}$ film with thickness 0.25 and 0.5 microns, and a dual layer Ni-Fe film with thickness of each layer 20 nm. The film was deposited on a cold substrate of polycrystalline silicon and fused silica. Experimental samples of films used in the research had the size 5x3 mm. Ni$_{81}$Fe$_{19}$ film can serve as a good model to study the environment of mutual features laser with metal films. Magnetic properties of such films are well understood. They have a high permeability and low coercive force, but their coercive force Hc and magnetic susceptibility pretty much depend on the structure and internal stresses.

Magnetic characteristics and hysteresis loop were measured with a vibration magnetometer. Measurements carried out on the microstructure of the electron microscope, analysis of results was carried out measurements on the X-ray and Auger spectrometer. The structure of the film surface was studied using atomic force microscope.

Experimental samples of the films were irradiated by nanosecond pulse of Nd:YAG laser ($\lambda$=1064 nm, $\tau$=30 ns and $\lambda$=355 nm, $\tau$=15 ns) with Gaussian distribution of energy in cross section and pulses of excimer laser ($\lambda$=248 nm, $\tau$= 20 ns). Uniformity of the intensity distribution in the cross section of the beam excimer laser was achieved with a special homogenizer. The stability of power was about 5%. Irradiation of films energy in the laser pulse was obviously less than the energy required to melt the film. The temperature of the film in the exposure was calculated and preliminary measurements of film damage threshold were carried out.

The results showed that roughness of film surface after irradiation of the laser pulses was significantly reduced. Moreover, such changes of roughness are observed after irradiation with laser pulses, the intensity of which is one and a half to two times less of the damage threshold (melting) of the film (Figure 1).

![Figure 1: Change in the roughness of surface of Ni$_{81}$Fe$_{19}$ film after irradiation with one pulse Nd:YAG laser (top, spherical lens) and excimer laser (bottom sequence of three pulses focused by a cylindrical lens).](image)
with one pulse Nd-YAG laser (\(\lambda=1064\) nm) and excimer laser focused pulse cylindrical lens (\(\lambda=248\) nm). The irradiation of excimer laser sample was moved after each pulse at 250 \(\mu\). The surface of the film after irradiation becomes smoother, and the loop region with a low roughness section coincides with the contour of the laser beam on the surface.

Interesting changes of the surface (Fig. 2) were observed after irradiating pulses of the films of the third harmonic of Nd: YAG laser (\(\lambda=355\) nm, \(\tau=15\) ns) with Gaussian distribution of energy in cross section. On the surface of the film structure at a high radiation intensity a little less film damage threshold is observed.

Figure 2. Photos of the film surface after irradiation pulses of Nd:YAG laser (\(\lambda=355\) nm): 1 – the sequence of single pulses, the laser beam diameter is 3-5 microns; 2 – the result of destruction of the film, the diameter of the laser beam was less than 1 micron.

Laser radiation reduces not only roughness of the surface, but also produces a useful change of magnetic characteristics of the films. The switching curve of the films becomes more rectangular and narrow after irradiation by even one single laser pulse (Figure 3). Besides, magnetic permeability \(\mu\) and the coercive force of the film \(H_c\) are changed. At a low energy density of the laser pulse (less than \(W=0,1\) J/cm\(^2\)) the film characteristics change little. With the increased energy density magnetic permeability \(\mu\) increases and coercive force of the film \(H_c\) decreases. For the pulses with a high energy density (on the damage threshold of the film), these magnetic properties change less.

The results showed that there is an optimal energy in the laser pulse, at which the magnetic film characteristics vary the most. However, the maximum amount of change in the magnetic characteristics of the films depends on the laser photon energy. For the film of 500 nm thick at magnetic susceptibility \(\mu\) for a Nd:YAG laser at \(\lambda=1064\) nm increased almost 3 times, and the coercive force \(H_c\) decreased about two.

Figure 3. Change of the form of the switching curve (I), and also magnetic permeability \(\mu\) and coercive force \(H_c\) (II) in film Ni\(_{81}\)Fe\(_{19}\) with the thickness of 500 nm after an irradiation nanosecond laser pulses: I-1 – switching curve loop of magnetic reversal to an irradiation, I-2 – pulse of Nd:YAG laser (\(\lambda=1064\) nm), \(W=0,6\) J/cm\(^2\); I-3 – pulse of excimer laser \(W=0,6\) J/cm\(^2\); II-1 – pulse of Nd:YAG laser (\(\lambda=1064\) nm) at different density of energy, II-2 – pulse of excimer laser \(W=0,6\) J/cm\(^2\).

For the excimer laser at \(\lambda=248\) nm, we have received an increase in the magnetic...
susceptibility $\mu$ almost ten times while a decrease in the coercive force $H_c$ was 5-6 times. Similar changes in the magnetic characteristics (Figure 4-I) are observed in the films irradiated by pulses of the third harmonic Nd:YAG laser ($\lambda=355$ nm, $\tau=15$ ns).

We also carried out the study of the effect of the initial temperature of the film on the changes of the magnetic characteristics under irradiating it with laser pulses. The samples had the same initial magnetic film parameters. Each sample was heated to temperature from 200°C to 400°C and irradiated with an excimer laser pulse energy density $W=0.6$ J/cm$^2$. The results of changes in the magnetic characteristics of different samples after irradiation pulse excimer laser are shown in Figure 4-II.

We observed analogous changes of magnetic characteristics in films Ni$_{81}$Fe$_{19}$ with thickness of 250 nanometers after an irradiation laser pulses. Distinction in change of magnetic characteristics was that the size of the maximum change of a magnetic susceptibility and coercive forces was in such films more than the maximum size change of similar characteristics in films Ni$_{81}$Fe$_{19}$ with thickness of 500 nm (Figure 5).

We conducted the study of the structure and composition of the film prior to irradiating and after irradiating it by the pulse excimer laser. Studies have shown that after irradiation with laser pulses the film composition does not change substantially, and only the structural changes occur in the surface layer. Figure 6 is a micrograph of the cross section of the film thickness of 500 nm.
Ni$_{81}$Fe$_{19}$ before irradiating and after irradiating it by the pulse excimer laser ($\lambda = 248$ nm and $\tau = 20$ ns) at a density of light energy $W = 0.6$ J/cm$^2$. It can be seen that, after irradiation, an increase in the size of the nanocrystals from the surface region of the laser pulse is observed. The value of such nanocrystals reaches 200-259 nm. We were not able to fix significant changes in the crystal structure after irradiation pulse Nd-YAG ($\lambda = 1064$ nm) laser. For the only samples we were able to pay, the changes in the crystal structure were little visible.

Figure 6. Change of crystal structure of Ni$_{81}$Fe$_{19}$ film with a thickness of 500 nm after an irradiation one nanosecond pulse of excimer laser with density of energy $W = 0.6$ J/cm$^2$: structure of cross-section section of the film to irradiation (1); structure of cross-section section of the film after irradiation excimer laser pulse with the density of energy $W = 0.6$ J/cm$^2$ (2).

It should be noted that on the damage threshold of film a regular structure appers after irradiation of 500 nm thick Ni$_{81}$Fe$_{19}$ uniform (Gaussian) beam of Nd-YAG laser ($\lambda = 355$ nm and $\tau = 15$ ns) on the verge of destruction (melt) film on the surface of the film (Figure 2, 1). The typical size of the structure element is about 200 nm, which makes it possible to connect it with a process of forming large nanocrystals.

We also studied the influence of nanosecond laser pulses on diffusion processes. Figure 7 shows the results of measuring the distribution of atoms in the two-layer Ni-Fe film before and after the irradiation with excimer laser pulse with the energy density $W = 0.6$ J/cm$^2$. The film is heated under such energy of pulse to the temperature of less than 1000$^\circ$ C.

Figure 7. Distribution of atoms in the two-layer Ni-Fe film: 1-Ni, 2-Fe; I - non-irradiated film, II - the film irradiated by the excimer laser pulse with the energy density $W = 0.6$ J/cm$^2$ from the nickel layer, III - the film irradiated by the excimer laser pulse with the energy density $W = 0.6$ J/cm$^2$ from the iron layer.

The measurement results show that under the influence of the laser pulse observed the asymmetry in the diffusion of iron and nickel atoms. The diffusion of atoms in the direction of the laser pulse is much stronger.
3. Discussion of the results and conclusions

Significant decrease of the roughness of surface shows that we have a melting or evaporation of needlelike nanoparticle on the surface under laser pulse. And this is when the radiation energy in the laser pulse is almost half of the energy that causes visible destruction of the surface of the film Ni$_{81}$Fe$_{19}$. This fact shows that we have a strong increase of the absorption of light or useful increase of the laser radiation field on such a needlelike nanoparticle of surfaces. This mechanism can be excitation of localized plasmon on the needlelike nanoparticle on the film surface, which results in the increase absorption and amplification of the electromagnetic field. A plane wave does not excite the surface plasmons on the flat top of the film, as the photon momentum is less than the momentum of surface plasmon. However, the localized plasmons on metal nanoparticles are excited by light of any polarisation. These plasmons increase absorption of light and the intensity of electromagnetic field of light wave near to the surface [3].

The field $E$ of the light wave excite in nanosphere with dielectric permeability $\mu$ a wave of polarisation $P$ [3].

$$P = E R^3 \frac{\varepsilon_1 - \varepsilon_0}{\varepsilon_1 + 2\varepsilon_0}, \ (1)$$

where $\varepsilon_1=\varepsilon_0(1-1/L)$, $L$ – geometrical shielding factor? $R$ – radius of curvature of the sphere, $\varepsilon_0$ – dielectric capacitivy in a medium.

We have a resonant excitation of particles when the term of fraction tents to zero. Such case is realized in metal particles at the expense of great value of the factor of absorption (the big size of an imaginary part of dielectric capacitivy $\varepsilon_{12}$) [4,5]. At an estimation of the size of amplification coefficient of electromagnetic field it is necessary to consider geometrical shielding factor $L$ for the given metal particle. This leads to the change in resonance conditions.

The size of amplification coefficient $\eta$ of electromagnetic field can be estimated on the basis of following expression

$$\eta = \left( \frac{E_1}{E_0} \right)^2 = \left( \frac{E_0}{3L^2\varepsilon_{12}} \right)^2, \ (2)$$

Where $E_1$ and $E_0$ – intensity of the field of incident wave and excited wave.

For an ellipsoid with eccentricity 3 it is equal $L=0.1$, and we will receive an useful increase of the size of amplification coefficient $\eta$.

In the field of falling of a laser beam on the film surface there is a big number of needlelike nanoparticles. All these needlelike nanoparticles are excited by laser radiation coherently. As a result, there is a considerable strengthening of the electromagnetic field of laser radiation at the expense of folding weaving excitation of all particles.

For two particles on distance $r$ amplification coefficient $\eta$ will register as

$$\eta = \left| \frac{B}{1-\alpha_1\alpha_2A^2} \right|^2 \left( \alpha_1 + 2\alpha_1\alpha_2A + \alpha_2 \right)^2 \ (3)$$

Where $\alpha=R^2(\varepsilon_{12} - \varepsilon_0)/3(\varepsilon_0 + L_1(\varepsilon_{12} - \varepsilon_0))$; $A=(r^2+ikr^2)e^{ikr}$; $B=2(8r^2+4kr^2)e^{-ikr}$.

This expression shows that amplification coefficient $\eta$ is much bigger then 1 ($\eta>>1$).

If two particles contact, the expression for amplification coefficient is possible to be written down as [5,6].
In this case amplification coefficient $\eta$ for 2 particles is in 30-50 times more than for one particle. At the big intensity of laser radiation it is necessary to consider the nonlinear response in polarizability of particles that will lead to a big increase of amplification coefficient $\eta$.

All stated above shows that localized on the needlelike nanoparticle plasmons play important role in processes of interaction of laser radiation with the surface of metal films. Such localized plasmons are a source of increase of factor of absorption and cause the amplification of a field of laser radiation falling on the film surface. These processes allow to reduce roughness of a surface of the film by means of laser radiation and thus there is no destruction on the film surface.

Our researches show, that not thermal processes play also the important role at the change of magnetic characteristics of $\text{Ni}_{81}\text{Fe}_{19}$ films under the action of laser pulses. That confirmation of this is quite a strong dependence of the efficiency of these processes from the laser photon energy. Laser pulse heating the film also influences the efficiency of the polycrystalline structure and changes the magnetic characteristics. We also believe that the effectiveness of the recrystallization depends on the degree of ionization of the atoms. Laser photon pressure also affects the efficiency of the process. This effect creates a film-depth nonequilibrium charge electrons [7. 8], which interact with the ionized atoms.

Since the depth of the recrystallization of the film coincides quite well with the depth of penetration of the laser radiation in the near-surface layer of the $\text{Ni}_{81}\text{Fe}_{19}$ film. The measured values of the absorption coefficient equal in $\text{Ni}_{81}\text{Fe}_{19}$ film for the excimer laser ($\lambda=248$ nm) at about $k=(3-6)\times10^4 \text{ cm}^{-1}$ and for the Nd:YAG laser ($\lambda=1064$ nm) $k=(3-5)\times10^5 \text{ cm}^{-1}$. The photon energy of the excimer laser is more than three times bigger of the photon energy of Nd:YAG laser ($\lambda=1064$ nm). Therefore, the thickness of the ionized layer of $\text{Ni}_{81}\text{Fe}_{19}$ film and the degree of ionization of atoms in it for the excimer laser is much greater than the thickness of ionized layer and the degree of ionization of atoms in it for the Nd:YAG laser. We believe that the speed and efficiency of the recrystallization depends strongly on the degree of ionization.

The effect of photon pressure is one of the factors that can significantly influence on the processes of recrystallization in the ionized layer. Under the laser-induced effect of photon pressure excited electrons move deep into the in the $\text{Ni}_{81}\text{Fe}_{19}$ film. The path length $l$ to such electrons is about $l=\frac{\hbar k}{2\pi m_e^*} \tau$. Where $\hbar k/2\pi$ and $\tau$ - photon momentum and time of laser pulse, $m_e^*$ - electron effective mass in the magnetic layer. The nonequilibrium charge of the electrons in the depth of the film may influence on the mobility of the ionized atoms and change the speed of recrystallization in the ionized layer of the $\text{Ni}_{81}\text{Fe}_{19}$ film under laser pulse.

The significant changes in the magnetic characteristics of the film under the influence of laser pulses shows that by using tightly focused short laser pulses can be generated in the polycrystalline magnetic NiFe films the regular structure of nanoislands.

References


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