Giant injection magnetoresistance induced by femtosecond laser pulses in semiconductor / granular film heterostructures with cobalt nanoparticles

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Abstract. The light-induced giant injection magnetoresistance in SiO₂(Co)/GaAs heterostructures, where the SiO₂(Co) structure is the granular SiO₂ film with Co nanoparticles, has been studied. It is found that the photocurrent are accompanied by relaxation oscillations caused by the Coulomb influence and transitions between the photocurrent and electrons on the highest level in the interface quantum well. The light-induced magnetoresistance reaches its maximum value in the avalanche onset region and has the local minimum at the higher voltage. The local minimum is explained by delocalization of the highest level in the interface quantum well and by decrease of the probability of the backscattering process of injected electrons on deeper levels.

1. Introduction
Ferromagnet-semiconductor heterostructures are of great interest because of promising applications for spin-electronic devices and magnetic sensors. SiO₂(Co)/GaAs heterostructure, where the SiO₂(Co) structure is the granular SiO₂ film with Co nanoparticles, is perspective material, in which the magnetoresistance reaches 1000 (10⁵ %) at room temperature [1-3]. Since the effect is expressed when electrons are injected from the granular film into the semiconductor, this magnetoresistance effect was called the giant injection magnetoresistance (IMR effect).

High values of the IMR effect in SiO₂(Co)/GaAs heterostructures are explained by the theoretical model of a magnetic-field-controlled avalanche process provided by electrons passed through the spin-dependent potential barrier in the accumulation layer at the interface and by the spin-dependent current reduction caused by the backscattering process of injected electrons on exchange-splitted levels of the interface quantum well [2-4].

Although high values of the magnetoresistance in SiO₂(Co)/GaAs heterostructures have been obtained, relaxation processes accompanying the IMR effect has not been studied. This study can clarify the nature of transitions occurring in the interface region of SiO₂(Co)/GaAs heterostructures and is important for production of fast-response magnetic sensors with higher sensitivity in comparison with GMR-sensors. In this paper we study dynamics of the giant injection magnetoresistance in SiO₂(Co)/GaAs heterostructures induced by femtosecond light pulses and investigate features of electron transitions occurring in the SiO₂(Co)/GaAs interface region.

2. Sample preparation and characteristics of SiO₂(Co) films
The (SiO₂)₁₀₀₋ₓCoₓ (or shorter SiO₂(Co)) films with the thickness of 81–95 nm were prepared by the ion-beam deposition technique using a composite cobalt-quartz target onto GaAs substrates heated to
200°C [2]. The Co concentration in SiO2 matrix was specified by a proportion of cobalt-quartz surface areas. n-GaAs substrates with thickness of 0.4 mm were of the (100)-orientation type. The substrate electrical resistivity was equal to 0.93×10^5 Ω·cm. Prior to the deposition process, substrates were polished by a low-energy oxygen ion beam [5]. The roughness height of the polished surfaces was less than 0.5 nm.

The film composition was determined by the nuclear physical methods of element analysis [6]. The cobalt to silicon atomic ratio in the samples (SiO2)_{100-x}Co_{x}/GaAs was measured by the Rutherford backscattering spectrometry of deuterons. For the samples studied, the relative content of cobalt x are in the range of 38 - 82 at.%. The average size of Co particles was determined by the small-angle X-ray scattering and increased as the concentration of x grows: from 2.7 nm at x = 38 at.% to 4.4 nm at x = 82 at.%. A protective Au layer with the thickness of 3-5 nm was deposited on the SiO2(Co) films. The Au layer formed one ohmic contact in experiments, while the second contact was on the GaAs substrate. Electrical resistivity of SiO2(Co) films was measured by the dc four-probe method on SiO2(Co)/quartz heterostructures at room temperature. As the Co content increased, the resistivity of SiO2(Co) films decreased from 1.46×10^6 Ω·cm (38 at.%) to 1.1 Ω·cm (82 at.%). The film resistivity is much smaller than the resistivity of the GaAs. In this case, in experiments the applied voltage U primarily falls on the GaAs substrates.

It was found that SiO2(Co) films sputtered on GaAs are inhomogeneous through the thickness. Small-angle scattering of synchrotron radiation in the grazing geometry (GISAXS) and X-ray reflectivity showed a specific interface layer 70-75 Å thick separating bulk SiO2(Co) granular film from the semiconductor substrate [7]. This layer was formed by a monolayer of flattened Co particles which were laterally spaced apart much further than the particles in the bulk film. From field dependencies of magnetization it was found that SiO2(Co) films are fully saturated for an applied field of 2 kOe for the parallel orientation. With the field perpendicular to the film, the value of 9 kOe is necessary to reach the saturation.

3. Injection magnetoresistance and relaxation processes induced by femtosecond pulses of light

Current-voltage characteristics and magnetoresistance in SiO2(Co)/GaAs heterostructures performed at direct current shows that the IMR dependencies are similar for samples with different cobalt concentrations. The only difference is in the location of the IMR coefficient maximum. Taking this into account, in order to study dynamics and relaxation processes induced by femtosecond light pulses we have chosen the SiO2(Co 60 at.%)/GaAs heterostructure. For this purpose, we used the circularly-polarized laser pulses with the duration of 180 fs and with photon energies of 1.35 eV and 1.4 eV. The pulse repetition was of 5 kHz and the light fluence was equal to 5 mJ/cm². The magnetic field was normal to the SiO2(Co) film surface (figure 1). The light beam has been passing through the Au contact layer, the SiO2(Co) film and the GaAs substrate. It is found that the highest growth of the photocurrent exists in the narrow range of photon energies of 1.38-1.41 eV near the GaAs bandgap energy $E_g$ [8]. Relaxation processes induced by femtosecond pulses of light were detected by microwave frequency band oscillograph. Parameters of the low-pass $RC$-filter have been chosen such that $RC > 10$ ms.
Relaxation process of the current flowing in the SiO₂(Co 60 at.%)/GaAs heterostructure induced by pulses of light at different applied voltages in the magnetic field \( H = 12 \text{ kOe} \) and without a magnetic field at the photon energy 1.35 eV is shown in figure 2. In the start region of the relaxation process relaxation oscillations are observed. After the light pulse, at the time \( t_2 \), the photo-induced current reaches its maximum value \( j_2 \). At the time \( t > t_2 \) the photo-induced current exponentially decreases. One can observe that the magnetic field suppresses the current. The current suppression reaches its maximum at \( t = t_2 \). It is found that within the experimental error, the photo-induced currents do not depend on the sign of the light helicity.

![Figure 2](image_url)

**Figure 2.** Relaxation process of the current flowing in the SiO₂(Co 60 at.%)/GaAs heterostructure induced by pulse of light at different applied voltages in the magnetic field \( H = 12 \text{ kOe} \) (curves 30 V (H), 60 V (H), 100 V (H)) and without a magnetic field (curves 30 V (0), 60 V (0), 100 V (0)) at the photon energy 1.35 eV. (a) Large time scale, (b) start region of the relaxation process. The curve fitting is presented for the 60 V (0) dependence.

The injection magnetoresistance induced by femtosecond pulses of light is defined as

\[
IMR^{(ph)} = \frac{j_2(0) - j_2(H)}{j_2(H)}
\]

where \( j_2(H) \) and \( j_3(0) \) are the photo-induced current at the time \( t_2 \) in the magnetic field \( H \) and without a magnetic field, respectively. The injection magnetoresistance coefficient \( IMR^{(ph)} \) (figure 3) reaches the maximum value in the avalanche onset region \( (U = 60 \text{ V}) \) in the GaAs and is equal to zero at the voltage \( U = 80 \text{ V} \).

![Figure 3](image_url)

**Figure 3.** The injection magnetoresistance \( IMR^{(ph)} \) versus the applied voltage \( U \) measured at the time \( t_2 \). Arrows mark minimum and maximum values of the injection magnetoresistance.
4. Discussion

Explanation of the experiment is based on the theoretical model developed in [2-4]. In SiO$_2$(Co)/GaAs heterostructures the difference of chemical potentials between the SiO$_2$(Co) film and the GaAs determines bending of the semiconductor conduction band (figure 4) and forms a quantum well (accumulation electron layer) in the semiconductor near the interface. Due to the exchange interaction between electrons in the quantum well in the semiconductor and $d$-electrons of Co, the quantum well contains exchange-splitted levels.

![Figure 4. Electronic energy band structure and exchange-splitted levels in the quantum well at the interface region in the granular film SiO$_2$(Co)/GaAs heterostructure. 1 and 2 mark exchange-splitted levels.](image)

The light impulse generates additional electrons in the conduction band and induces a photocurrent. The dynamics of the photocurrent caused by these additional electrons and variations of densities of electrons on exchange-splitted levels 1 and 2 can be determined by equation

$$\frac{\partial}{\partial t} \begin{pmatrix} n_j \\ n_1 \\ n_2 \end{pmatrix} = D \begin{pmatrix} n_j \\ n_1 \\ n_2 \end{pmatrix} + \begin{pmatrix} A \delta(t) \\ 0 \\ 0 \end{pmatrix},$$

where $n_j$ is the density of electrons induced by light. The photocurrent is determined by $n_j$, $j = en_j v$, where $e$ is the electron charge and $v$ is the electron velocity. $n_1$ is the variation of the density of electrons on the level 1 (first level) in comparison with the thermodynamic equilibrium density. By analogy, $n_2$ is the variation of the density of electrons on the level 2 (second level). The matrix $D$ is

$$D = \begin{pmatrix} -(a_1 + a_2) & (b_1 - V_1) & (b_2 - V_2) \\ a_1 & -(c + b_1) & 0 \\ a_2 & c & -b_2 \end{pmatrix},$$

where $a_1$ and $a_2$ are the probabilities of transitions of photocurrent electrons on the first and second levels per unit of time, respectively. $V_1$ and $V_2$ determine the Coulomb influence of electrons localized on the first and second levels, respectively, on the photocurrent $j$. Increase of electrons on the levels leads to a decrease of the current $j$. $b_1$ and $b_2$ are the probabilities of electron tunneling per unit of time through the barrier from the levels 1 and 2, respectively. $c$ is the probability of the electron transition per unit of time from the first level on the second level. The term with $A \delta(t)$ in equation (1) denotes the generation of the photocurrent by the light. We take into account that the pulse width of light is to a great extent less than the observed relaxation times. $A$ is the coefficient.

The eigenvalues $\lambda^{(k)}$ are determined by equation

$$\det(D - \lambda^{(k)} I) = 0,$$

where $I$ is the identity matrix. In the experiment we observe relaxation oscillations, consequently, one of the eigenvalues must be complex ($\lambda^{(1)} = \text{Re} \lambda^{(1)} + i \text{Im} \lambda^{(1)}$, $\text{Re} \lambda^{(1)} \neq 0$, and $\text{Im} \lambda^{(1)} \neq 0$). The second eigenvalue $\lambda^{(2)}$ is real. Since the probability $b_1$ of electron tunneling from the first level is to a great extent higher than the probability $b_2$ of electron tunneling from the deeper second level and taking into account that the probabilities of transitions on the deeper second level are smaller than the
probabilities of transitions on the higher first level \((c < b_1, a_2 < a_1)\), from equation (2) one can approximately calculate the relaxation time \(\tau_1\) and the circular frequency of relaxation oscillations

\[
\tau_1 = \frac{-1}{\text{Re} \lambda^{(1)}} = \frac{2}{a_1 + b_1},
\]

\[
\omega = \text{Im} \lambda^{(1)} = \frac{1}{2} [4a_1V_i - (a_1 + b_1)^2]^{1/2}
\]

(3)

In this case, we take into account only interaction between the photocurrent and the variation of the electron density \(n_1\). Influence of the second level on the photocurrent \(j\) and, consequently, the term \((b_2 - V_2)\) in the \(D\)-matrix (1) are ignored. Relaxation oscillations are observed, if the Coulomb term satisfies the inequality

\[
V_i > \frac{(a_1 + b_1)^2}{4a_1}.
\]

In our experimental study we observe relaxation oscillations with the frequency of \(\omega/2\pi = 100\) MHz. Within the experimental error the relaxation oscillation frequency \(\omega/2\pi\) is not varied under the magnetic field action and with increase of the applied voltage \(U\). The relaxation of the photocurrent \(j(t)\) can be written as

\[
j(t) = -B_1 \exp \left(-\frac{t}{\tau_1}\right) \cos(\omega t) + B_2 \exp \left(-\frac{t}{\tau_2}\right),
\]

(4)

where \(B_1\) and \(B_2\) are coefficients connected with the current \(j_1\) (figure 2) by relation \(j_1 = B_2 - B_1\). In order to find the relaxation times \(\tau_1\) and \(\tau_2\), we have fitted experimental relaxation curves of the photocurrent (figure 2) by the theoretical dependence (4) with \(\omega = 0\).

The relaxation time \(\tau_1\) of the photo-induced current flowing in the \(\text{SiO}_2(\text{Co 60 at.\%})/\text{GaAs}\) heterostructure in the magnetic field \(H = 12\) kOe and without a magnetic field at the photon energy 1.35 eV is presented in figure 5. One can see that within the experimental error the relaxation time \(\tau_1\) does not depend on the magnetic field. At high values of the applied voltage \(U > 90\) V the small decrease of the relaxation time \(\tau_1\) is observed. According to relation (3), in the main, the relaxation time \(\tau_1\) is determined by transitions and the Coulomb influence between the photocurrent and electrons on the first level.

\[
\begin{array}{c}
\text{Figure 5. Relaxation time } \tau_1 \text{ of the photo-induced current flowing in the } \text{SiO}_2(\text{Co 60 at.\%})/\text{GaAs} \text{ heterostructure in the magnetic field } H = 12 \text{ kOe (} *, \text{ curve 1.35 eV(H)) and without a magnetic field (} *, \text{ curve 1.35 eV(0)) at the photon energy 1.35 eV.}
\end{array}
\]

\[
\begin{array}{c}
\text{Figure 6. Relaxation time } \tau_2 \text{ of the photo-induced current flowing in the } \text{SiO}_2(\text{Co 60 at.\%})/\text{GaAs} \text{ heterostructure in the magnetic field } H = 12 \text{ kOe (curves 1.35 eV(H) and 1.4 eV(H)) and without a magnetic field (curve 1.35 eV(0)) at photon energies 1.35 and 1.4 eV.}
\end{array}
\]
Dependencies of the relaxation time $\tau_2$ of the photo-induced current flowing in the same heterostructure in the magnetic field $H = 12$ kOe and without a magnetic field at photon energies 1.35 and 1.4 eV versus the voltage $U$ is presented in figure 6. It should be noted that the voltage growth results in significant decrease of the relaxation time $\tau_2$. Besides, in the magnetic field the relaxation time $\tau_2$ increases. In contrast with the relaxation time $\tau_1$, high values of the time $\tau_2$ is caused by electron transitions occurred on the deeper second level.

The above-mentioned model of the dynamics of the photocurrent with exchange-split levels in the interface quantum well can explain the zero value of the injection magnetoresistance coefficient IMR$_{\text{ph}}$ (figure 3) at the voltage $U = 80$ V. Indeed, voltage growing bends the semiconductor conduction band. As a result of the bending, the higher first level (figure 4) can be delocalized. In this case, the first level drops out from the photocurrent dynamics model and, besides the second level, it is necessary to take into consideration deeper levels in the quantum well. The probability of the backscattering process of injected electrons on deep levels is small [3]. This leads to small variations of the density of electrons on these levels, and, therefore, to low values of the magnetoresistance IMR$_{\text{ph}}$. The further growth of the voltage results in increase of bending of the conduction band and the subsequent level lifting. As a result, the IMR$_{\text{ph}}$ coefficient increases.

As a result of the performed study, one can conclude that SiO$_2$(Co)/GaAs heterostructures can be used as efficient fast-response magnetic sensors operating at room temperature. The upper frequency bound of sensors is of 1-5 MHz.

5. Conclusion

Relaxation processes of the photocurrent in SiO$_2$(Co)/GaAs heterostructures induced by femtosecond pulses of light are accompanied by relaxation oscillations. Relaxation oscillations are caused by the Coulomb influence and transitions between the photocurrent and electrons on the highest level of exchange-split levels in the interface quantum well.

The light-induced magnetoresistance IMR$_{\text{ph}}$ reaches its maximum value in the avalanche onset region and has the local minimum at the higher voltage. The local minimum of the IMR$_{\text{ph}}$ dependence is caused by delocalization of the highest level in the interface quantum well and by decrease of the probability of the backscattering process of injected electrons on deeper levels.

Acknowledgments

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