Journal of Magnetism and Magnetic Materials xxx (2017) xxx–xxx

Contents lists available at ScienceDirect

Journal of Magnetism and Magnetic Materials

journal homepage: www.elsevier.com/locate/jmmm

Research articles

Magneto-optical spectroscopy of diluted magnetic semiconductors GaMnAs prepared by ion implantation and further impulse laser annealing

E.A. Gan'shina [a,*], L.L. Golik [b], Z.E. Kun'kova [b], G.S. Zykov [a], I.V. Bykov [c], A.I. Rukovishnikov [b], Ye Yuan [d], R. Böttger [d], S. Zhou [d]

[a] Department of Physics, Lomonosov Moscow State University, Moscow, Russia
[b] Kotel'nikov Institute of Radioengineering and Electronics (Fryazino Branch), RAS, Fryazino, Russia
[c] Institute for Theoretical and Applied Electromagnetics, RAS, Moscow, Russia
[d] Helmholtz-Zentrum Dresden Rossendorf, Institute of ion-beam science and Materials Research, Dresden, Germany

ARTICLE INFO

Article history:
Received 10 July 2017
Received in revised form 17 October 2017
Accepted 14 November 2017
Available online xxxx

Keywords:
GaMnA
Magneto-optics
Ellipsometry
Ion implantation
Laser annealing

ABSTRACT

$Ga_{1-x}Mn_xAs$ layers prepared by ion implantation and subsequent pulsed laser annealing with the planned Mn concentrations of $x = 0.01–0.08$ have been studied using the magneto-optical transversal Kerr effect (TKE) and spectral ellipsometry. The spectral dependences of the diagonal and nondiagonal components of the permittivity tensor (PT), as well as the spectrum of magnetic circular dichroism (MCD) have been calculated for the layers. The obtained spectra of the diagonal PT components show that the layers under study maintain the zinc-blende crystal structure of the parent GaAs semiconductor. All studied samples reveal a strong TKE response at low temperatures with a dependence of an effective Curie temperature (at which TKE appears) on the Mn concentration. A number of extrema in the low-temperature TKE and MCD spectra are close to the energies of transitions in the Γ and L critical points of the parent semiconductor band structure that confirms the intrinsic ferromagnetism of the $Ga_{1-x}Mn_xAs$ layers. The MCD spectra shape and its change with Mn concentration increasing are discussed on a base of the valence-band model of ferromagnetism in DMS.

1. Introduction

The (Ga,Mn)As diluted magnetic semiconductor (DMS) is the model object both for ascertainment of the nature of ferromagnetic coupling in the (III,Mn)V DMS and for control of their magnetic properties. However, despite the intensive research the debates regarding the electronic states near the top of the valence band and at the Fermi energy as well as the exchange mechanisms leading to ferromagnetic ordering still continue [1–3]. Some authors assume that in heavily doped ferromagnetic (Ga,Mn)As the Fermi level lies in the narrow impurity band separated by an energy gap from the valence band, and the metallic conductivity of such samples is due to the hole motion along the impurity band (the IB-hole model) [4–8]. At the same time, according to [9–12], there is no the impurity band detached from the valence band in ferromagnetic metallic (Ga,Mn)As samples, and the Fermi level is located in the valence band (the VB-hole model).

In parallel with probing the spectrum of electronic states magneto-optical (MO) spectroscopy allows one to detect and identify various magnetic phases, which can be present in (Ga,Mn)As samples. Magnetic circular dichroism (MCD) is usually considerably enhanced around the critical points of semiconductor band structure [13], therefore MCD spectroscopy is especially widely exploited. However, the experimental MCD data available in the literature are interpreted by authors from different research groups as evidences supporting both points of view.

Low-temperature molecular beam epitaxy (LT-MBE) is the most common way to obtain (III,Mn)V systems [14,15] and the majority of studies has been performed on LT-MBE (Ga,Mn)As samples. In the course of the low-temperature growth the point defects (interstitial Mn and antisite As) arise, which, being donors, compensate largely the majority carriers (holes) and reduce the ferromagnetic ordering temperature, T_C, of (Ga,Mn)As. The discrepancies between the experimental results of different authors, apparently, are related to the problem of fabrication of the samples with well-controlled defects.

* Corresponding author.
E-mail address: eagan@mail.ru (E.A. Gan'shina).

https://doi.org/10.1016/j.jmmm.2017.11.058
0304-8853/© 2017 Published by Elsevier B.V.

The other growth techniques such as metal organic vapour phase epitaxy (MOVPE) [16], ion implantation and subsequent pulsed laser annealing [17,18] as well as pulsed laser deposition [19] are also used to prepare (Ga,Mn)As layers. In the case of these methods, other defect types may arise during the growth process that can have a significant impact on the character of impurity states, concentration of itinerant holes and the Fermi level location. Therefore, studies of (Ga,Mn)As grown by the alternative methods can provide additional information on the details of the (Ga,Mn)As electronic structure, particularly about the Fermi level position.

In our previous studies [20–22] the MO technique (the transversal Kerr effect, TKE) was used together with spectral ellipsometry to characterize the IIIMnV layers (III = Ga, In; V = As) grown by laser deposition. It has been ascertained that these layers contain ferromagnetic MnAs inclusions, which appreciably determine the layers properties, in particular their strong MO response at room temperature. In the TKE spectra of such layers both the structures associated with the optical transitions in "bulk" MnAs inclusions and the resonant TKE band (E = 0.5–2.0 eV) conditioned by excitation of the surface plasmons can be present. The knowledge about these spectra features allows one to detect the presence of the undesirable MnAs phase in Ga(In)MnAs layers.

In this paper, we present results of the investigations of magneto-optical and optical properties of GaMnAs layers fabricated by ion implantation and subsequent pulsed laser annealing. The results show that the MnAs phase is absent in our samples. The samples demonstrate ferromagnetic behaviour at low temperatures. We have calculated spectra of the real and imaginary parts, $\varepsilon'_1(E)$ and $\varepsilon\varepsilon'_2(E)$, of the nondiagonal components of the permittivity tensor as well as spectra of the magnetic circular dichroism for samples under study and compared them with the published MCD spectra of (Ga,Mn)As grown by LT-MBE [5,6,10,23–26]. Features in the MCD and TKE spectra of our samples and their change with increasing Mn concentration are explained on a base of the valence-band model of ferromagnetism in DMS, taking into account the electronic phase separation and quantum size effect.

2. Material and methods

2.1. Ion implantation and further impulse laser annealing

The $Ga_{1-x}Mn_xAs$ layers on GaAs(0 0 1) substrates were prepared by ion implantation at room temperature at the Ion Beam Center at the Helmholtz-Zentrum Dresden-Rossendorf. Mn ions were implanted into intrinsic GaAs(0 0 1) wafers at the energy of 100 keV at an incidence angle of 7° to avoid ion channeling. According to SRIM simulations, the Mn concentration was planned to be from 0.01 to 0.08 considering fluence of 1×10^{15}–2×10^{16} cm^{-2}. A coherent XeCl excimer laser with 308 nm wavelength and 28 ns duration was used to anneal the implanted GaAs layer. The profile of the laser beam was homogenized to 5×5 mm^2. The annealing energy was optimized to be 0.3 J/cm^2. However, the laser annealing made the amount of implanted Mn atoms (inside the GaAs layer) diverged from the planned value, because part of the implanted Mn atoms diffused to the surface during the recrystallization procedure. Later on, the sample surface was etched in concentrated HCl to remove the inert surface layer. Nevertheless, substantial part of the Mn atoms still remained inside the GaAs matrix. The Mn concentration was measured by secondary ions mass spectrometry (SIMS) and it was determined by the obtained peak value. Fabrication conditions, x values planned and determined by SIMS, samples spontaneous magnetization, M_s, and thickness as well as Curie temperature magnitudes, T_C, got from magnetometry, magneto-optical and electrical measurements are presented in Table 1. Magnetometry was performed using a SQUID-VSM magnetometer from Quantum Design. More details about the samples fabrication are shown in Ref. [17,18]. Data of SIMS and cross-sectional high-resolution TEM as well as of magnetometry and electrical measurements for the given samples are presented in our work [27].

2.2. Magneto-optical and optical spectroscopy

Magneto-optical transversal Kerr effect, TKE, consists in an intensity variation of the p-polarized light reflected by a sample under magnetization. The value $\delta = [I(H) - I(-H)]/2I(0)$, where $I(H)$ and $I(0)$ are the reflected light intensities in the presence and absence of a magnetic field, respectively, was directly measured in the experiment. The alternating magnetic field up to 240 kA/m was aligned parallel to the sample surface and perpendicular to the light incidence plane. The sensitivity of the apparatus was $\approx 10^{-5}$. The TKE spectra, $\delta(E)$, were recorded in the energy range of $E = 0.5 - 4.0$ eV. The temperature range for the studies was $T = 15 - 300$ K. The TKE value depends linearly on magnetization and we have measured temperature dependences, $\delta(T)$, as well as TKE dependences on magnetic field, $\delta(H)$, at some fixed energies to characterize magnetic state of the layers.

To record the ellipsometry parameters spectra, $\tan\psi(E)$ and $\cos\Delta(E)$, we employed the ellipsometer SE 850 DUV SENTECH Instruments GmbH (the energy range of $E = 0.55 - 6.5$ eV, samples #1 – #3, #6,) and ellipsometer with binary modulation of light polarization state [28] ($E = 1.24 - 4.5$ eV, samples #4 and #5). The spectra were recorded at room temperature.

3. Results

3.1. Spectral ellipsometry

We used the ellipsometry spectra, $\tan\psi(E)$ and $\cos\Delta(E)$, to derive the spectral dependences of the real and imaginary parts, $<\varepsilon_1>$ and $<\varepsilon_2>$, respectively, of the diagonal components of the pseudo-dielectric function[1], for the samples investigated. The obtained $<\varepsilon_1>(E)$ and $<\varepsilon_1>(E)$ spectra are shown in Fig. 1. Dashed lines show the energies of the transitions at the Γ- and L- critical points of GaAs at room temperature [30]. As is clear from Fig. 1, peaks corresponding to the optical transitions near the L critical point of the Brillouin zone of GaAs are present in the $<\varepsilon_2>(E)$ spectrum of all $Ga_{1-x}Mn_xAs$ samples. Therefore, we can conclude that the crystal structure of the GaAs parent semiconductor is conserved in the layers under study. An increase in matrix imperfection when the Mn dopant is introduced is a possible reason for decreasing the maxima and smearing the doublet ($E \approx 2.9 - 3.2$ eV), as well as for the increase in the $<\varepsilon_2>$ values in the region $E \approx 1.2 - 2.9$ eV. The transformation of the $<\varepsilon_1>(E)$ and $<\varepsilon_2>(E)$ dependences in the energy region $E < 3$ eV is also associated with the light interference in the $Ga_{1-x}Mn_xAs$ layers. The interference contribution appears and rises below the E_1 transition energy, where the optical absorption in the semiconducting matrix abruptly decreases and the depth of the light penetration into the sample increases. The interference fringes are especially well marked in the spectra of sample #6. In Fig. 1 arrows denote these fringes.

A possible reason for the interference in sample #6 can be its relatively high transmission in the range $E < 3$ eV. Note that the structures associated with the transitions E_1 and $E_1 + \Delta_1$ in the spectra of this sample are smeared weaker than those in the other samples spectra. This is apparently due to smaller imperfection of

[1] The pseudo-dielectric function is obtained directly from the measured $\tan\psi$ and $\cos\Delta$ values with the use of the optical model assuming a flat substrate with infinite thickness [29].

Please cite this article in press as: E.A. Gan'shina et al., Magneto-optical spectroscopy of diluted magnetic semiconductors GaMnAs prepared by ion implantation and further impulse laser annealing, Journal of Magnetism and Magnetic Materials (2017), https://doi.org/10.1016/j.jmmm.2017.11.058