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Forming Interface in Pd/Fe/GaAs/InGaAs Structure for Optical Detector of Free-Electron Spin

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Abstract—Conditions necessary for the formation of a Fe/GaAs interface have been established and the electrical, magnetic, and optical properties of Pd/Fe/GaAs heterostructures with InGaAs quantum wells have been studied. The possibility of obtaining an epitaxial layer of Fe on GaAs(001) surface at room temperature is demonstrated. The magnetization curve of Fe layer exhibits hysteresis with an easy axis in plane of the sample. Iron exhibits surface segregation by diffusion through a 4-nm-thick Pd layer. The properties of obtained Pd/Fe/GaAs/InGaAs structures show evidence for their possible use in optical detectors of free-electron spin.

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The growth of Fe/GaAs structures and their magnetic and transport properties have been extensively studied in the past decade in view of the potential possibility of using ferromagnet/semiconductor structures in spintronics [1, 2]. One possible application of these structures is a free-electron spin detector [3, 4]. This detector can be implemented, e.g., based on a Pd/Fe/GaAs(001) heterostructure with a magnetic Schottky barrier capable of measuring the spin polarized current of a beam of free electrons in vacuum, which can also be used as a spin-polarized electron injector in solid-state spintronics [5]. The magnetic Schottky barrier in a ferromagnet/semiconductor structure plays the role of a spin filter that predominantly transmits electrons with the spin collinear with the magnetization vector of the ferromagnetic layer. By measuring the difference between the injected spin-polarized electron current for the opposite directions of film magnetization or electron spin, it is possible to measure the spin polarization of the injected electron beam. The main difficulty that is encountered in manufacturing the required ferromagnet/semiconductor structure is related to the creation of a ferromagnet/GaAs(100) structure with a sufficiently low density of defects and surface states, since the spin detector under consideration requires a magnetic Schottky barrier with a relatively large area (~0.5 cm^2) with a low (~10^{-7} A/cm^2) leakage current density.

An alternative approach to measuring the spin polarization of an electron beam is offered by the optical method that consists in detecting cathodoluminescence in the single-photon regime and measuring the degree of polarization of the recombination radiation. The use of the optical detection method significantly decreases the requirements imposed on the electrical properties of the ferromagnet/GaAs(100) interface. This Letter presents the results of an investigation of the electrical, magnetic, and optical properties of Pd/Fe/GaAs heterostructures with InGaAs quantum wells (QWs), which are intended for the creation of optical detectors of free-electron spin.

The sample structures were manufactured by two-stage epitaxial growth technology. At the first stage, GaAs structures with two $In_{0.18}Ga_{0.82}As$ quantum wells were grown by molecular beam epitaxy (MBE) in a Riber 32 setup. For this purpose, a 100-nm-thick GaAs buffer layer doped with Si up to $n \sim 10^{18}$ cm^{-3} was formed on an n-GaAs(001) substrate. This was followed by p-type ([Be] ~ 7×10^{17} cm^{-3}) AlGaAs and GaAs layers with thicknesses of 40 and 20 nm, respectively (deposited at $T = 580$°C), and two 10-nm-thick p-$In_{0.18}Ga_{0.82}As$ quantum wells separated by a 20-nm-thick p-GaAs layer (grown at $T = 480$°C). The upper, 20-nm-thick undoped GaAs layer was grown at the same temperature. At the second stage, metal (Fe and Pd) layers were deposited.

Heterostructures based on the Fe/GaAs system involved the interface between Fe and a clean GaAs(001) surface with $(4 \times 2)/c(8 \times 2)$ reconstruction. The reconstructed GaAs(001) surface was prepared by treating with HCl solution in isopropyl alcohol (HCl–IPA), followed by heating in vacuum to $T =$ 550°C [6]. Then, metals (Fe, Pd) were deposited in vacuum by evaporation from Knudsen cells. The deposition rate was 0.1 nm/min. After the formation of a 4-nm-thick Fe layer, a 4-nm-thick Pd layer was deposited for protecting iron from oxidation in air. Both metals were deposited at room temperature and a residual gas pressure in the technological chamber not exceeding 1×10^{-9} Torr.

The structure of Pd/Fe/GaAs heteroepitaxial films was studied by high-resolution transmission electron microscopy (TEM) on a JEOL-4000EX instrument (Japan) operating at an accelerating voltage of 400 kV. The samples were prepared as transverse sections using the standard method of mechanical polishing followed by ion-sputter thinning.

The chemical composition of layers in the heterostructure was studied by X-ray photoelectron spectroscopy (XPS) on a SPECS spectrometer (Germany) equipped with a PHOIBOS-150-MCD-9 energy analyzer and a FOCUS-500 X-ray monochromator using AlK_α radiation with $h\nu$ =1486.74 eV at an X-ray tube power of 200 W. The scale of binding energies (E_b) was calibrated relative to the positions of core electron levels Au4$f_{7/2}$ (84.00 eV) and Cu2$p_{3/2}$ (932.67 eV). The electron binding energy and full width at half maximum (FWHM) of the photoelectron peaks were measured accurate to within 0.05 eV. The ion etching was performed by a beam of 1.2-keV argon ions generated by an IQE 11/35 ion gun at a current density of 2 μm/cm^2. The residual pressure in the analyzer chamber did not exceed 5×10^{-5} Torr.

Photoluminescence (PL) measurements were performed at room temperature using solid-state lasers operating at different wavelengths (λ = 375, 532, and 660 nm). Figure 1a presents a schematic diagram of the Pd/Fe/GaAs/InGaAs heterostructure and shows the geometry of measurements. The emission was excited from the side of epitaxial layers via ferromagnetic metal layers, and the PL spectra were measured from the substrate side. An analogous scheme is typically used for the detection of cathodoluminescence [7].

Figure 1b shows a TEM image of the Pd/Fe/GaAs structure cross section in the (110) plane. As can be clearly seen, the layers of Pd and Fe possess a crystalline structure, which is evidence for the epitaxial deposition of metals onto the GaAs(001) surface. The epitaxial growth of a Fe film on GaAs(001) is related to the fact that the lattice parameter of iron amounts to almost exactly half of that for GaAs (0.5653 nm), which implies a relatively small lattice misfit parameter (~1.4%) [8]. During the epitaxial growth of the cubic metal lattice on the cubic semiconductor lattice, the spatial frequency of the Fe lattice in the (001) plane is doubled as compared to that of GaAs. The possibility of epitaxial growth of Fe on GaAs at both low (<100°C) and high (>300°C) temperatures on reconstructed surfaces was demonstrated in [9]. Investigation of the sample surface by atomic force microscopy (AFM) showed evidence of an insignificant growth in the surface roughness after deposition of the metal layers: the root-mean-square (rms) roughness height (determined on a 1×1 μm^2 area) increased from 0.15 nm for the initial GaAs(001) surface to 0.3 nm for the metal-coated one.

Figures 1c and 1d present the X-ray photoelectron spectra of a Pd(4 nm)/Fe(4 nm)/GaAs(001) structure measured (*1*) after one-year storage in air and (*2*) after removal of a 2-nm-thick outer metal layer by argon ion etching. The Pd 3*d* spectra of the initial surface reveal a shoulder at ~337.2 eV on the high-energy side, which is evidence for the presence of oxidized palladium on the sample surface [10]. The oxide layer thickness, calculated from the ratio of intensities of the line of bulk Pd to that of Pd in the oxygen environment (with allowance for a photoelectron mean free path of λ = 1.8 nm), corresponded to about two monolayers (~0.3 nm).

Analysis of the Fe 2*p* spectrum shows that it consists of two peaks, the most intense of which occurs at a binding energy of 711 eV, which corresponds to iron in the state of Fe^{3+} ions occurring in an oxygen environment, i.e., in the form of Fe_2O_3 oxide. The less intense peak has a binding energy of 707 eV that corresponds to Fe–Fe bonds of elemental iron in the bulk. A low intensity of the bulk iron component is related to screening of the iron layer by the upper layer of palladium. The presence of oxidized iron on the surface of palladium is evidence for the surface segregation of iron. The amount of iron at the surface corresponds to several monolayers.

As can be seen from curves *2* in Figs. 1c and 1d, argon ion etching of the sample leads to the removal of both palladium and iron oxides from the surface. Indeed, the bonding energy for iron amounts to 707.0 eV and corresponds to the Fe–Fe bonds of elemental iron. For the Pd 3*d* level, the thinning of the uppermost layer leads to a shift of this line to a higher binding energy of ~336.2 eV, which is ~1 eV higher than the value (335.1 eV) characteristic of the bulk metal [11]. The shift of the Pd 3*d* line by up to 0.6 eV toward higher binding energies in the presence of iron was reported in [12]. It can be suggested that the segregation and, probably, mixing of iron with palladium lead to a significant change in the electron structure of palladium.

Thus, an analysis of the chemical composition of the Pd/Fe/GaAs structure shows that the growth of Pd/Fe layers at room temperature is accompanied by the segregation of iron and its subsequent oxidation on the surface of palladium. However, despite the segregation and partial mutual solubility of iron and palla-

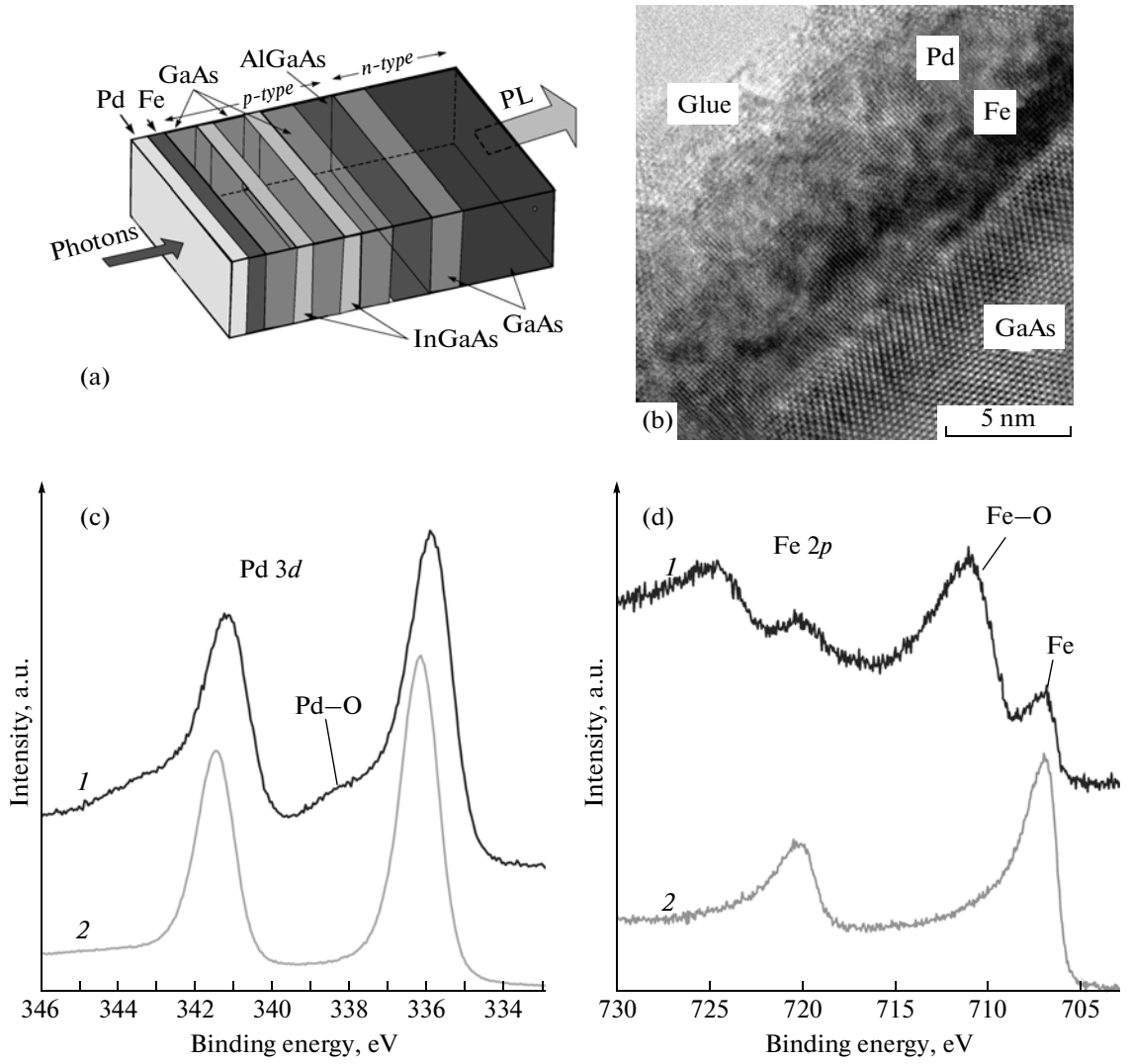

Fig. 1. Pd/Fe/GaAs(001) heterostructure: (a) schematic diagram of layer sequence and geometry of PL measurements; (b) TEM image of interfaces in the cross section; (c, d) X-ray photoelectron spectra of Pd 3*d* and Fe 2*p* levels, respectuively, on the surface of Pd(4 nm)/Fe(4 nm)/GaAs(001) structure measured (*1*) after one-year storage in air and (*2*) after removal of 2-nm-thick outer metal layer by argon ion etching.

dium, the magnetization of iron in this system corresponds to that of bulk iron (with allowance for the layer thickness) [4]. The retained magnetization of iron is probably explained by the formation of a ferromagnetic state in the PdFe alloy [13].

Figure 2 shows the current–voltage (I–V) characteristic of the Pd/Fe/GaAs/InGaAs heterostructure, which describes the current passage through a p–n junction. In contrast to the magnetic Schottky barriers [4], quality requirements to the Fe/GaAs(001) interface for the optical detection of electrons injected into semiconductor are not as rigorous. The inset to Fig. 2 presents a curve of the iron film magnetization in the surface plane measured using the magneto-optical Kerr effect (MOKE). Magnetization of the Fe/GaAs(001) structure was always characterized by a rectangular hysteresis curve, an easy axis oriented along the crystallographic direction [110], and a coercive field of about 25 G. On the whole, the magnetization and coercive field values weakly changed depending on the angle of sample rotation in the plane. The presence of a hysteresis confirms the homogeneity (monodomain character) of the film and is determined by the epitaxial growth of Fe on GaAs(001). It should be noted that the interface between iron and GaAs(001) surface covered with oxides is characterized by a significant anisotropy of the magnetization [4].

In experiments on the measurement of spin-dependent cathodoluminescence, emission will be detected from the substrate side (in the transmission mode). Therefore, it is important to study luminescent properties of the proposed Pd/Fe/GaAs/InGaAs heterostructure with respect to the light transmitted

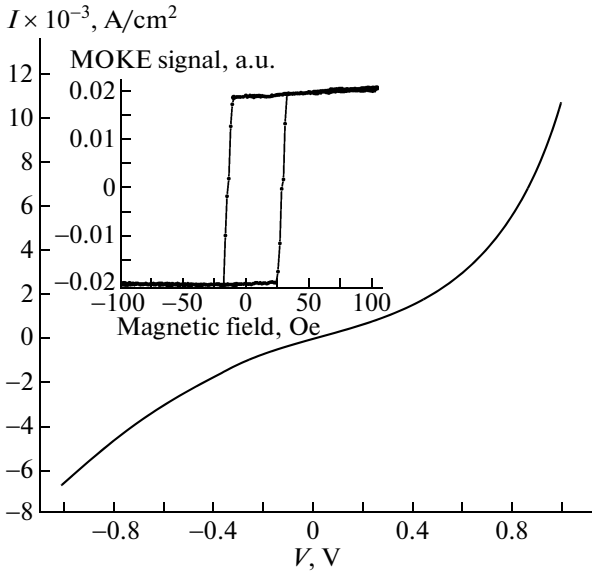

Fig. 2. Current–voltage (I–V) characteristic and (inset) magnetization curve of the Pd/Fe/GaAs/InGaAs heterostructure.

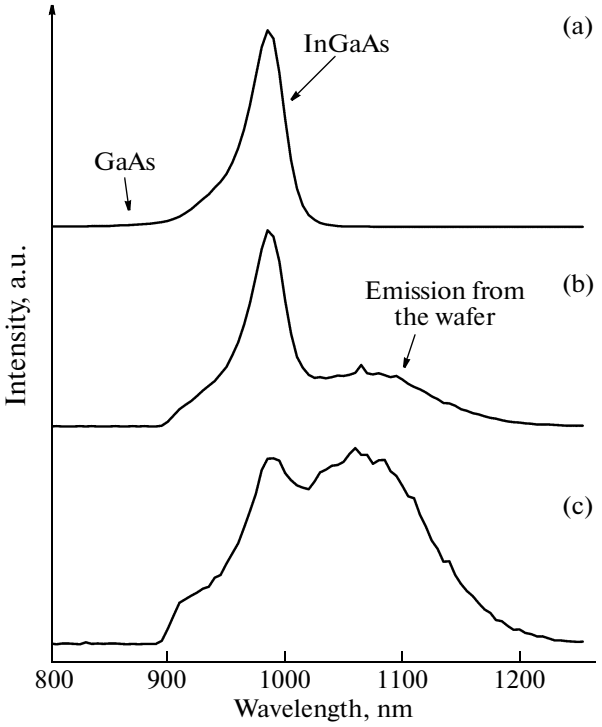

Fig. 3. PL spectra of a Pd/Fe/GaAs/InGaAs heterostructure illuminated from the metallization side by laser radiation with wavelengths (λ = 375 (a), 532 (b), and 660 nm (c). The emission was detected from the GaAs substrate side. The measurements were performed at T = 300 K.

through the structure. Figure 3a presents the PL spectrum measured from the substrate side for the structure excited from the metallization side by UV laser radiation with a wavelength of λ = 375 nm ($h\nu$ = 3.30 eV). This PL spectrum displays a single peak with a maximum at λ = 980 nm, which corresponds to the recombination emission from InGaAs quantum wells. Under illumination by the light with λ = 532 nm (2.33 eV), the PL spectrum exhibits an additional broad band in the region of 1050–1200 nm (Fig. 3b). This emission is due to a recombination via the levels of impurity–defect complexes (IDCs) present in the *n*-GaAs substrate. In contrast to the case of a UV light, which is completely absorbed near QWs, the radiation with λ = 532 nm has a greater depth of absorption and partly penetrates into the substrate, thus generating electron–hole pairs that recombine via IDCs. A contribution from the substrate to the PL spectrum is even more clearly pronounced in the case of excitation by laser radiation with λ = 660 nm (1.88 eV) that corresponds to an even greater depth of absorption (Fig. 3c). Note that a peak due to the edge luminescence from GaAs (λ = 870 nm) is absent in the observed spectra (Fig. 3) because of a strong absorption of this emission by the substrate. The peak of GaAs edge luminescence was observed when the PL was measured from the metallization side.

Thus, our investigation of the electrical, magnetic, and optical properties of Pd/Fe/GaAs heterostructures with InGaAs quantum wells showed the possibility of measuring cathodoluminescence in the transmission mode and applying these structures in optical detectors of free-electron spin.

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