Nanocrystalline Magnetic Semiconductors for Spintronics

Subjects: Physics, Condensed Matter

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Challenge for spintronics is to find new ways for the control of electronic phenomena and magnetic properties of solids at nanoscale. Some promising aspects are connected with developing new materials. The research highlights the areas devoted to the creation of new functional materials for spintronics based on magnetic semiconductors and demonstrates the technical possibilities of creating various devices, in particular, a maser, a p-n junction with a colossal magnetoresistance, a spin valve, a magnetic lens, modulators, spin wave amplifier, etc. A magnetic semiconductor is a magnetic material that, in terms of specific conductivity, occupies an intermediate position between a conductor and an insulator, and has a band gap comparable to ~*kBT*. Most known magnetic semiconductors (SC) are either oxides or chalcogenides (sulfides, selenides and tellurides) of 3d transition metals, rare earth 4f metals or a combination. Spintronics (spin electronics) studies spin current transfer (spin-polarized transport) in condensed media, including contact structures, heterostructures, superlattices and multilayers. Much attention is paid to the mechano-physical methods of obtaining of high-density transparent nanoceramics based on magnetic semiconductors. The potential possibility of using nanoceramics as an absorber of solar energy, as well as in modulators of electromagnetic radiation, is also presented. The THz magneto-optics in magnetic semiconductors is shown to be beneficial to the intensively developing fields of spintronics – ultrafast magnetooptics and magnetophotonics in magnetic semiconductors.

Keywords: spintronics ; magnetic semiconductors ; complex magnetic oxides

1. Introduction

A magnetic semiconductor is a magnetic material that, in terms of specific conductivity, occupies an intermediate position between a conductor and an insulator, and has a band gap comparable to ~k_BT. Most known magnetic semiconductors (SC) are either oxides or chalcogenides (sulfides, selenides and tellurides) of 3d transition metals, rare earth 4f metals or a combination. The strong interaction of mobile charge carriers with the localized magnetic moments of the d- and f-shells leads to a number of features of the electrical and optical properties of magnetic SC that are absent in non-magnetic semiconductor [1][2][3][4][5][6][7][8][9].

There are several properties typical only for magnetic SC. For example, in ferromagnetic semiconductors (europium chalcogenides, chromium chalcogenide spinel, etc.), with either a decrease in temperature, application of an external magnetic field or both, a giant (up to ~0.5 eV) shift towards long wavelengths of the edge of optical absorption and photoconductivity is observed, the so-called giant red shift of the absorption edge. Meanwhile, near the Curie temperature they can exhibit a colossal magnetoresistance (CMR) effect and a metal-insulator (MI) transition. Giant magneto-optical (MO) effects were found in magnetic SCs in a wide visible and infrared (IR) spectral range (Faraday and Kerr effects, magnetotransmission, magnetoreflection, magnetic linear and circular dichroism, etc.) [10][11][12][13][14]. Moreover, magnetic SCs are the basis of high-temperature superconductors and without considering the properties of magnetic semiconductors, it would be impossible to construct a theory of high temperature superconductivity (HTSC). By changing the concentration of current carriers (by temperature, doping, light irradiation, application of electric field, current injection, etc.), it is possible to easily change the magnetic properties of SCs [1][2][3][4][5][6][7][15][16][17][18][19][20]. On the contrary, their electrical and optical properties can be controlled by an external magnetic field, which in relation to the intense development of electronics and magnetophotonics is of high practical interest [21][22][23][24][25]. For example, the presence of a large magnetocaloric effect in manganites makes magnetic SCs promising materials for creating refrigerators [26]. Large MO effects and the presence of the MI transition in magnetic oxides made it possible to create a wide range of optoelectronic devices, such as modulators, magnetic field visualization sensors, MO filters, shutters, attenuators, bolometers, thermochromic materials, etc. [27][28][29][30][31][32][33]. The electrical properties of perovskites and cobaltites allowed for using them as cathodes in solid oxide fuel cells, opening new possibilities for creating alternative energy

sources with a high efficiency of ~90% [34][35]. The medical direction of using magnetic SCs is also very important, for example, as a material for artificial local heating of a selected area of the body-hyperthermia [36][37][38][39][40].

Unlike non-magnetics, magnetic semiconductors have the important property of dependence of the band structure on the orientation of the electron spin. In other words, in magnetic SCs, the band gap is not the same for electrons with different spin orientations (**Figure 1**).

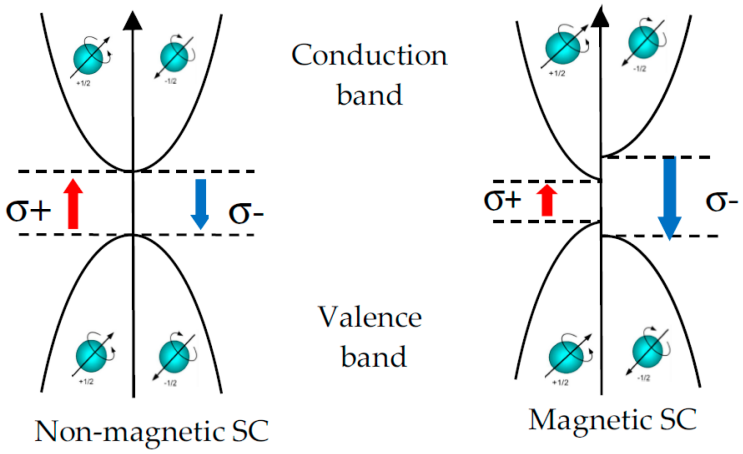

Figure 1. Schematic representation of changes in the band structure of a magnetic SCs depending on the orientation of the electron spin (**right**), which are absent in the band structure of a non-magnetic semiconductor (**left**).

The presence of bands with different orientations of the of charge carriers' spins (spin-polarized bands) made it possible to control the charge current in magnetic SCs, depending on the mutual orientation of the magnetic field, current direction and an applied voltage. It is important that the flow of spin-oriented (spin-polarized) charged particles retains its properties not only in magnetic SCs, but also in other magnetic or non-magnetic media at a sufficiently level. In non-magnetic semiconductors, spins with different orientations compensate each other, thus the full spin current is zero and there is only the current of charged particles. The possibility of creating and manipulating a flow of spin-polarized charge carriers in magnetic SCs has opened broad prospects for creating new functional materials and various electronic devices. Spintronics (spin electronics) studies spin current transfer (spin-polarized transport) in condensed media, including contact structures, heterostructures, superlattices and multilayers. In all these cases, the source of spin-polarized electrons (spin injector) is a ferromagnet (metal or semiconductor), which in the magnetized state has the spontaneous spin ordering of the current carriers [15][16][17][18][19][20][21][22][23][24][25]. In ferromagnetic SCs, the levels of spin polarization can reach higher values (up to almost 100%) than in metals (up to 10%) [41]. In non-magnetic SCs in an external magnetic field, the Zeeman splitting of the conduction band occurs with the formation of two energy sublevels. When spin-polarized electrons are injected from a magnetic SC into a non-magnetic SC, controlled transitions to both the upper and lower energy levels become possible, which allows controlling the charge current by an external influence. This area of physics has been intensively developed over the past 30 years [42][43][44][45][46][47][48][49].

At the same time, current spintronic devices or their components are generally extremely small (~10^{-7} m) and can be classified as nanomaterials. When the dimensions of device components or a material are reduced to a nanometer scale (for example, films, thin-film structures, fibers, powders and high-density ceramics), their mechanical properties are changed significantly, thermal conductivity is increased and a change of fundamental characteristics such as the melting temperature and Debye temperature were also reported. In a magnetic SC, during the transition to the nanoscale, the electronic structure is drastically changed followed by the appearance of various anomalies in its magnetic and transport properties [50][51].

2. Nanocrystalline Magnetic Semiconductors—New Functional Materials for Spintronics

Nanocrystalline magnetic SCs are of interest both for fundamental problems of condensed matter physics and for practical applications, due to the appearance of new properties, when the particle size of a SC is reduced to the nanometer scale [52][53][54]. For the past decade the development of magnetic materials, particularly manganites and ferrites in nano-form, has undergone a fast evolutionary growth as strategic materials in spintronics and sensor applications [55][56][57][58]. At the same time, the concentration of defects in nanomaterials is much higher than in equilibrium single or polycrystals. This factor also significantly determines the properties of nanomaterials, transforming nanosized magnetic SCs into multifunctional materials. The features and the possibility of using nanosized ferromagnetic SCs as functional materials

are well studied. Meanwhile, similar features are also manifested in nanosized antiferromagnetic SCs. For example, the presence of linear dichroism (~50%) allows for the use of antiferromagnetic semiconductor CuO as polarizers of infrared radiation [59]. Electron-field emission in CuO nanofibers is recommended for use in displays [60]. Irradiated nanocrystalline CuO oxides can accelerate reaction-catalytic effects in the production, for example, of alcohols. Moreover, the synthesis rate, for example, of copper phthalocyanine, can be controlled by a magnetic field at room temperature [61][62]. Nanocrystalline CuO is used in electronics as a buffer layer in complex thin-film electronic nanodevices [63]. Moreover, it can enhance an electrochemical activity, which is promising for use as electrodes in lithium current sources [64] and as an analyzer of carbon dioxide concentration [65]. However, there are problems in obtaining high-quality optically transparent SC nanomaterials of various types (powder, ceramic, fiber, and film).

2.1. High-Density Magneto-Optical Nanoceramics

For creating ceramics from a nanocrystalline material, important conditions are the preservation of the composition and size of compacted particles. Since magnetic SCs are multicomponent compounds, the production of high-density nanoceramics is rather complicated. To create nanoceramics, dynamic and static loading methods for obtaining high-density bulk nanomaterials are used. Static loading refers to the method of severe plastic deformation—shearing under pressure of at least 8 GPa. In this method, the number of distortions is determined not only by the applied high static pressure, but also by the angle of rotation of the anvils. For the creation of nanoceramics based on magnetic SCs, special high-pressure chambers were developed [66][67]. For example, using the static deformation method, $LaMnO_{3+y}$ nanoceramics with a density of 99% and grain sizes close to those of a nanopowder can be obtained [68]. It was also found that at high degrees of deformation and small crystallite sizes, the magnitude of micro-deformations in nanoceramics decreases, which may be due to more efficient relaxation of lattice stresses by grain boundary sliding with a decrease in crystallite size.

Dynamic loading refers to the method of explosive action on a low-density (polycrystalline powder compact) coarse-grained material made of a magnetic SC by spherically converging shock waves, as well as pressing axially-symmetric workpieces under the action of an explosion [69][70]. When exposed to spherically converging shock waves, the material is placed in a spherical steel hermetic case, in which, at the moment of a spherical explosion, the material is simultaneously compressed in all ways and a nanostructure is formed. During explosive pressing of axially-symmetric workpieces, the material is placed in a steel cylinder, in which, at the moment of explosion, the detonation wave propagates along the generatrix axis. The integrity of specially designed metal covers is not violated, which protects the resulting nanoceramics from contamination. The created nanoceramics show high temperature- and time-stability. As a result, the advantage of the developed methods is the ease of implementation, the combination of the creation of a nanostructure and material compaction in a single process, the production of high-density (~99%) stable nanosized materials and the absence of external contaminants. The results can be used for obtaining nanoceramics from a wide range of materials. Some fragments of real SC nanoceramics prepared by various methods are shown in **Figure 2**. It can be seen that the described methods allow obtaining dense nanomaterials of quite large geometrical sizes.

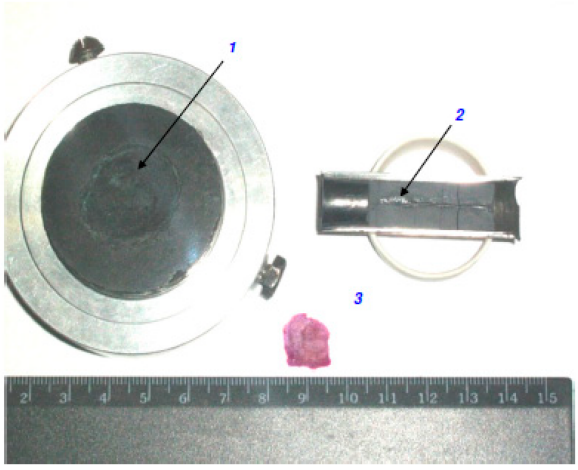

Figure 2. High-density nanoceramics obtained (1) by the method of spherically converging shock waves (CuO); (2) by the method of cylindrical explosive loading ($LaMnO_{3+y}$); (3) by the method of static pressure with shear ($Y_3Fe_5O_{12}$).

The promising technological process for creating high-quality dense nanoceramics of magnetic SC based on binary and even ternary compounds, such as CuO, Cu_2O, Mn_3O_0, ZrO_2, ZnSe, $LaMnO_{3+y}$, $FeBO_3$, $Y_3Fe_5O_{12}$, etc., has been developed at the Institute of Metal Physics UB of RAS, Russia. The average particle size varies from 10 to 100 nm. The crystal structure, microstructure, imperfection, microhardness, magnetic, optical and other properties of the obtained

nanomaterials have been studied in [71][72][73][74][75][76]. It has been established that the specific defectiveness of nanoceramics of magnetic oxides is ascribed to either the high concentration of oxygen vacancies, their agglomerates concentrated at the boundaries of crystallites or both, which determines (along with the nanostructure) the nonequilibrium nature of nanoceramics and the features of their mechano-chemical properties. For example, anomalies in the magnetic properties, a decrease of the forbidden band gap and a negative thermal expansion for nanoceramics were found.

In the creation of new functional nanomaterials, the production of transparent nanoceramics is very important. The aforementioned techniques made it possible to create transparent nanoceramics with a high Faraday effect, i.e., the rotation of the plane of polarization of linearly polarized light passing through nanoceramics under the influence of an external magnetic field.

2.2. Nanocrystalline $Y_3Fe_5O_{12}$ as a New Magneto-Optical Material

An optically transparent high-density (density 99.6%) $Y_3Fe_5O_{12}$ (YIG) nanoceramic (**Figure 2**) was obtained by means of a sheared static pressure of ~50 GPa. The authors of [77][78] showed that the absorption coefficient of nanoceramic $Y_3Fe_5O_{12}$ in the transparency window is below 50 cm^{-1}. It is mainly due to light scattering by crystallites and varies slightly with reduction of grain size. As the degree of deformation of YIG nanoceramics increases, the concentration of defects changes and the magnitude of micro-deformations decreases. However, these factors have only little effect on the absorption coefficient in the transparency window. **Figure 3** shows that the value of the Faraday rotation for nanoceramics of YIG is only 1.5 times less than that for the YIG single crystals and exceeds 50 deg/cm. It is important that as the grain size decreases, the Faraday effect increases, and then, starting from a grain size of 21 nm, the effect decreases again. The observed increase of the Faraday effect in nanosized magnetic SCs was theoretically explained in [79]. It was shown that electric dipole transitions in clusters of mixed valences of 3d ions in nanosized magnetic materials with the structure of perovskite and garnet can lead to a resonant enhancement of the MO activity in the limited area of deformations. Finally, transparent high-density nanoceramics based on magnetic SCs can be widely used for creation of various high-frequency and microwave sensors and shields and MO devices, in particular, IR radiation modulators, displays, etc.

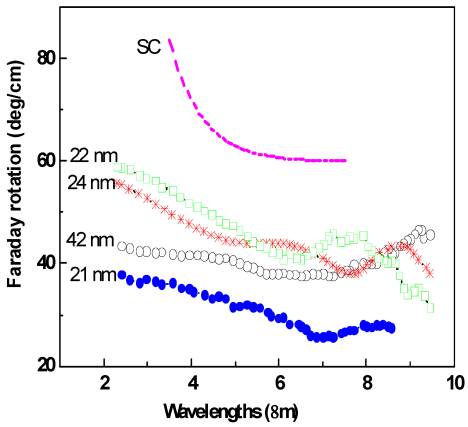

Figure 3. Specific Faraday rotation of nanostructured YIG samples with different crystallite sizes, SC–YIG single crystal.

2.3. Nanocrystalline CuO as a Material for Selective Solar Energy Absorbers

In relation to the problem of energy saving, great interest has arisen in alternative green energy sources, in particular, in selective absorbers of EM radiation and thermal converters of solar energy with coatings based on selective absorbers [80]. A selective solar energy absorber must meet certain requirements. For example, it should have a large absorption coefficient and a low reflection coefficient in the spectral range of solar radiation—in the energy range $E > 0.5$ eV. On the other hand, the absorber should have low electromagnetic energy emission or high reflectance at $E < 0.5$ eV. In other words, a selective absorber must absorb the energy of the radiation source as efficiently as possible but minimally re-emit it back into the atmosphere in the IR region of the spectrum. It was shown that copper oxides [81][82][83] can be used as selective absorbers in solar energy collectors with working temperatures up to T~500 °C. However, these materials do not meet the above conditions well, since they have a large forbidden gap. The authors of [84][85] demonstrated that this problem can be solved by changing the absorption spectrum of CuO practically without changing the refractive index when the copper oxide is transferred to the nanocrystalline state. **Figure 4** shows the optical density (absorption) spectra for CuO single-crystal, nanopowder and nanoceramic, respectively.

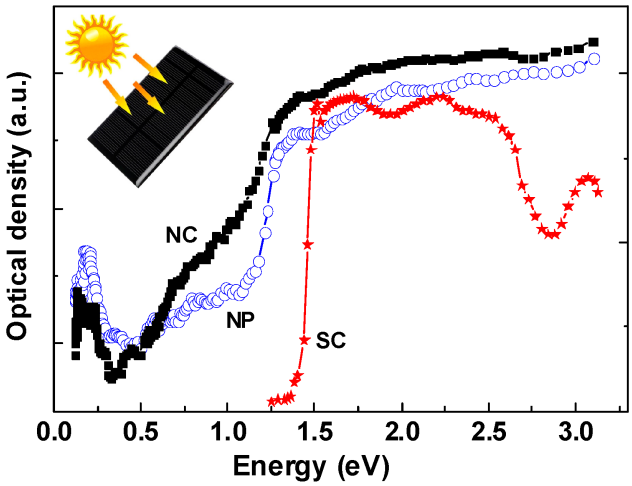

Figure 4. Optical density spectra of CuO samples at T = 295 K: SC—single crystal, NP—nanopowder, NC—nanoceramics.

The reasons for such strong changes in the spectra of CuO nanopowders and nanoceramics in the region of the main fundamental absorption edge were associated with a high level of defects and micro-deformations as well as with the behavior of strongly correlated materials in the nanocrystalline state. In CuO nanoceramics the observed energy changes that result in a significant decrease in the effective high-energy edge of the transparency window from 1.5 to 0.5 eV are the most pronounced [52][53].

It is necessary to note that there are many other directions in the development of technologies for obtaining nanoceramics and nanocomposites based on magnetic SCs and their possible applications in spintronics, nanophotonics, biology and medicine [86][87][88][89][90].

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