A short note on the organic semiconductors and their technical applications in Spintronics

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(Received 3 July 2013, accepted 30 November 2013)

Abstract

The organic semiconductors (OSCs) have been the topic of great interest in the research and development of the spintronic devices, after the initial progress and evolution of spintronics, as they have important applications in such systems. Some of the important technical applications of the OSCs in spintronics have been reviewed in this note. The behaviour of the electrical properties of the OSCs has been briefly discussed. The Mathematical analysis of the spin of an electron as the intrinsic angular momentum, and spin polarizer has also been presented.

Keywords: Organic Semiconductors (OSCs), Organic Tunneling Devices, Organic Magnetoresistive Devices.

Resumen

Los semiconductores orgánicos (OSC) han sido tema de gran interés en la investigación y el desarrollo de los dispositivos de espintrónica, tras los avances iniciales y la evolución de la espintrónica, ya que tienen aplicaciones importantes en este tipo de sistemas. Algunas de las aplicaciones técnicas importantes de las OSCs en espintrónica han sido revisadas en esta nota. El comportamiento de las propiedades eléctricas de las OSC se ha discutido brevemente. El análisis matemático del espín de un electrón como el momento angular intrínseco, y el polarizador giro también se ha presentado.

Palabras clave: Semiconductores organicos (OSCs), Dispositivos de tunelación orgánica, Dispositivos orgánicos magnetorresistivos.

PACS: 73.63.-b, 72.25.-b, 31.15.E-, 73.40.-c, **ISSN 1870-9095**

I. INTRODUCTION

An organic semiconductor is an organic material having semiconductor properties. The various available semiconductive forms are - single molecules, short chain (oligomers) and organic polymers. Organic semiconductors (OSCs) have become the topic of interest in the field of spintronics. Major breakthroughs have been achieved in their use as a tunnel barrier in magnetoresistive tunneling devices, and as a medium for spin-polarized current in transport devices. The technology based on organic semiconductors has made great strides in the last decade. Organic light-emitting diodes (OLEDs) have become commercially available and a lot of attention is being paid to using the organic photovoltaic (OPV) devices for some novel applications. The electric memories for non-volatile data storage and the organic field effect transistors (OFETs), have also been the topics of great activity in the research community. The OSCs have many fundamental advantages like low cost, lightweight, large area coverage. In addition, it is much easier to engineer their molecular properties apart from the mechanical flexibility compatible with the plastic substrates. The peculiar behaviour of the (OSCs) is due to the unusual properties of the carbon atom - like the configuration called sp2-hybridisations where the sp$_2$-orbitals form a triangle in a plane, and the pz-orbitals are in a plane perpendicular to it. An orbital overlap of two sp$_2$-orbitals leads to the formation of an s -bond between the two carbons. The energy difference between the occupied binding orbitals and the unoccupied anti-binding orbitals is quite large, and also beyond the visible spectral range. This means that the longer chains of bound carbon atoms have a large gap between the highest occupied molecular orbital (HOMO) and the lowest unoccupied molecular orbital (LUMO), which results in the insulating properties.

The Polymer organic semiconductors are formed due to the fact that when a long chain of carbon atoms is formed, the p -bonds become delocalized along the chain, and hence form a 1-D electronic system, which has considerable band width (~ 1eV). In this way, we have a 1 - D semiconductor with a filled valence band from the HOMOs and an empty conduction band from the LUMOs. The transport properties of such polymers are determined either by the defects in the 1D-chains or by hopping from chain to chain. It is important to note that the thermally activated hopping polymer organic semiconductors are deposited by wet process like spin-coating.

OSCs differ from inorganic counterparts in their optical, electronic, chemical and structural properties. For designing and modeling the OSCs, the optical properties like absorption and photoluminescence have to be characterized. The optical characterization can be done by using UV-visible absorption spectrophotometers and photoluminescence spectrometers. The film appearance and morphology are studied by using atomic force microscopy (AFM) and scanning electron microscopy (SEM). It is also necessary to characterize the electronic properties such as ionisation potential, which is done by studying the electronic band structure with ultraviolet photoelectron spectroscopy. The field of OSCs has thrown great challenges in the field of spintronics, mainly owing to the weakness of the spin-relaxation mechanisms [1]. As discussed [2], the spin polarization of the carriers is expected to be retained for very long times, which therefore, allows the spins to be manipulated. The use of OSCs has resulted in a new generation of spintronic devices with multiple functions, like sensing of magnetic, electrical, optical and chemical behaviours.

Spin injection in OSCs has already been observed in the studies [3] on the magnetoresistance (MR) of spin valve (SV) devices having OSCs layers of ~100–200nm thickness. Interesting and important studies have been made on OSCs in tunneling devices at the room temperature operation [4], and also with very high MR values ~ 600%.

Though a number of achievements have been made in the OSCs-based spintronic devices, they are found to suffer from a lot of scattering in the device performance, which shows that there are many limiting factors of both technological and fundamental nature, which have to be probed and understood. Some of these devices are - organic charge/spin-conducting spacer and inorganic ferromagnetic electrodes, the performance of which mainly depends on the properties of the materials used as the organic spacer and magnetic injecting electrodes, though it is also affected by the interfacial properties. It is necessary to understand the complex electric and magnetic phenomena taking place at these hybrid interfaces for designing and realizing the OSCs spintronic devices.

II. GENERAL CHARACTERISTICS OF THE ORGANIC SEMICONDUCTORS

In the inorganic semiconductors, the band structure is well established, and the significant carrier delocalization takes place by the combination of the covalent bonds and the structure symmetry. Interestingly, in the OSCs a strong intramolecular covalent carbon–carbon bonding combines with a weak van der Waals interaction between molecules to generate the materials with optical properties, which are quite similar to those of their constituent molecules, but with the transport properties governed and determined by the intermolecular interactions.

The electrical properties of OSCs usually result from the charge injection and charge transport. It should be noted that the charge injection from a metal into the OSC is quite different from the case of metals/inorganic semiconductors, due to the extremely low density of intrinsic carriers. The carriers propagate randomly by following a site-to-site hopping path between pseudo-localized states, and the drift direction is determined by an external electric field. Transport depends significantly on two factors: (i) the degree of order of the molecules, and (ii) the density of chemical and/or structural defects. It is seen that in OSCs, the impurities unlike in inorganic semiconductors, do not induce additional carriers, but lead to the generation of the trap states. The distributed shallow (~ below 0.1eV) and deep (about 0.5eV to 1eV) traps characterize the electrical properties of many OSCs, and it is noticed that their role is stronger in low-order materials like Alq3 polymers – Facial and Meridional Isomers of Tris(8-hydroxyquinolinate)aluminum [5]. This is precisely the reason that the experimental characterization of the transport properties in organic thin films or crystals is mainly determined by the sample quality.

It has been reported [6] in that the spin in an organic LED can now be measured, which is considered an important step toward a spintronic device. As has been discussed [7, 8], the spintronic devices are mostly made by using the magnetic metals, and also the conventional inorganic semiconductors like silicon and gallium arsenide. However, the use of the organic semiconductors has many advantages, since the organic devices can easily be made, as it is quite easy to deposit and structure them. Zhan and Fahlman [9] have presented the important results in organic spintronic research, particularly the spinterfaces, spin-injecting interfaces involving organic semiconductor (OSCs) molecules and ferromagnetic metals. The different categories of the interfaces depending on the type and strength of interface interaction, and the relevant physics underlying the energy level alignment and the spin polarization states of the interface have been discussed. They have studied and discussed in detail the models for spin injection at spinterfaces. In addition, the likely role of such devices on the device design and performance has been highlighted.

(i) ORGANIC TUNNELING DEVICES The research and development efforts on the devices involving spin-polarized tunneling across an organic barrier have so far been able to achieve quite high values of the magnetoresistance (MR), which are good enough for the sensor applications. Tunneling has been shown in nanopores with a self-assembled monolayer of octanethiol barrier [10] for devices with Langmuir–Blodgett monolayer barriers [11]. However, it has been observed that these results are not consistently reproducible, because they depend upon many parameters, which are not easy to control. The spin polarization of carriers is accurately measured by using the Meservey–Tedrow method [12]. The possibility of achieving spin-polarized tunneling across organic or organic–inorganic hybrid barriers with significant MR values [13] competitive with the case of the inorganic spintronics, has already been established**.**

(ii) SPIN SCATTERING IN ORGANIC SEMICONDUCTORS The dynamics by which OSCs modify the spin state of an electron are quite different, in the sense that, for the significant coupling strength, the spin of the carrier is not a good quantum number, but in fact, is a linear combination of spin-up and spin-down eigen states. In this manner, the spin-flip probability for the orbital momentum scattering events is defined by the OSC. Understanding of the case of the OSCs is quite difficult because of their completely different behaviour as far as the band conductivity versus random incoherent hopping is concerned. The tentative analyses of MR data on the basis of OSC scattering have been discussed in the studies [14, 15].

(iii) ORGANIC SPINTRONICS INHERENT MULTI-FUNCTIONALITY Devices based on organic spintronics have the great advantage that they can perform multiple functions. It is really interesting to note that the OSCs respond to the two or more independent electrical stimuli by giving an output or by acquiring a given state, which results in showing the resistive memory effects [16]. Also, the electrodes, on being applied by a magnetic field orient their magnetizations. Thus, it is clear that both these effects can be applied in an organic spintronic device [17].

The inherent Multi-functionality of the OSCs will lead to the development of future integrated memory-logic devices like the one for the electrical control of the MR, which is really very important. The spin metal oxide semiconductor field effect transistor is very important, as this is a 3-D device in which the gate can be used to turn on and off the SV effect between the drain and the source electrodes.

(iv) INTERFACES IN ORGANIC SPINTRONIC DEVICES All the spintronic devices (both tunneling and injection) based on the OSCs have a hybrid organic–inorganic interface for sustaining the spin-transfer effect. As is the case with the inorganic spintronic devices, the interface quality is crucial in the OSCs based devices also, which is really difficult to achieve. However, it is important to note that the behavior of the hybrid interfaces is quite intriguing, and still more work is required to fully understand it.

III. MATHEMATICAL ANALYSIS OF SPIN

The role of spin in the devices based on spntronics is important for studying (i) Spin of an Electron as the Intrinsic Angular Momentum, and (ii) II-Mn-VI/III-V Spin Polarizer.

(i) SPIN OF AN ELECTRON AS THE INTRINSIC ANGULAR MOMENTUM As is now understood, the spin of the electron is its intrinsic angular momentum, which is quite different from the angular momentum due to its orbital motion. The electron's spin is half of the reduced Planck constant which is equal to the Planck constant divided by 2π, and is denoted $\hbar$ (h-bar) *i.e.* $h/2\pi$, which implies that the electron acts as a Fermion by the spin-statistics theorem. The spin has orbital angular momentum, and also an associated magnetic moment, the magnitude of the later is given by:

$$m = \frac{\sqrt{3}}{2}\frac{\hbar q}{m_e}, \qquad (1),$$

where m is the angular momentum, m_e is the mass of the electron, and q is the charge of the electron.

Obviously, in a solid the spins of many electrons combine to affect the magnetic and electronic properties of the material *e.g.* produce a permanent magnetic moment in the material, as in a ferromagnet.

If, as is the case with many materials, electron spins are equally present in both - up and down states, then no transport properties are dependent on spin. It is necessary to generate or manipulate a spin-polarized population of electrons in a spintronic device, which results in an excess of electrons in either of these states. If X is any spin dependent property, then the polarization of X *i.e.* PX can be written as:

$$PX=(X_{up}-X_{down})/(X_{up}+X_{down}), \qquad (2),$$

where X_{up}, and X_{down} are respectively the spin up and spin down states of the electron. The net spin polarization is achieved by (i) creating an equilibrium energy splitting between spin up and spin down *e.g.* by putting a material in a large magnetic field (Zeeman effect), or (ii) the exchange energy present in a ferromagnet; or forcing the system out of equilibrium. The non-equilibrium spin population can be maintained for a period of time τ, which is the spin lifetime, The distance over which a non-equilibrium spin population can propagate is called the diffusion length λ. Another problem faced is that the spin lifetimes of conduction electrons in metals are relatively short ~ 1 nanosecond, and a great research effort is required to extend this lifetime to the practical levels suitable for the technological applications.

(ii) II-Mn-VI/III-V SPIN POLARIZER The case of II-Mn-VI/III-V Spin Polarizer is also very interesting, and it has been found that the optimum value of the polarization fits in the following equation:

$$P\,(optimum) = [(3n_{up} + n_{down}) - (3n_{down} + n_{up})] \,/\, [(3n_{up} + n_{down}) + (3n_{down}+n_{up})], \qquad (3)$$

where n_{up} and n_{down} are respectively the numbers of electrons with spin up and spin down states. For example, if the fraction of n_{up} is 0.7, and that of n_{down} is 0.3, then P (*optimum*) according to the Eq. (3) comes out to be equal to 0.2. Conversely for P (*optimum*) ~ 1, the following equation has to hold good:

$$[(3n_{up} + n_{down}) - (3n_{down} + n_{up})] = [(3n_{up} + n_{down}) + (3n_{down}+n_{up})] \quad (4).$$

This implies that, $2n_{up} - 2n_{down} = 4n_{up} + 4n_{down}$. *i.e.*

$$2n_{up} + 6n_{down} = 0. \quad (5)$$

Thus, for *P*(*optimum*) ~ unity, n_{up} has to be ~ 1/3rd of n_{down}. This has important implications for the engineers designing the II-Mn-VI/III-V Spin Polarizers**.**

(iii) II-VI DILUTE MAGNETIC SEMICONDUCTORS (DMSs) The 100% spin-aligner is developed by using the II-VI Semiconductors with magnetic components (e. g. ZnMnSe), which are non-magnetic or antiferromagnetically aligned at zero B-field. For finite magnetization, giant Zeeman-splitting of up to 100 meV (of which 10 - 20 meV is in the conduction band), has been observed. The other important observations are: (i) Low Fermi energy at high doping due to impurity bands, and (ii) Half-metallic behavior at high B-field. Spin injection is characterized by an 'interface resistance' that does the spin conversion, which can be measured by magnetizing the DMS.

IV. CONCLUSIONS

Spintronics is the branch which manipulates the electron spin (or resulting magnetism) to achieve the new or improved functionalities like spin transistors, memories, higher speed, lower power, tunable detectors and lasers, bits (Q-bits) for quantum computing. It has been established that in case of the Metal Oxide Semiconductor Field Effect Transistor MOSFET, the Gate Voltage changes electron density, which further changes conductivity. In case of the spin transistor, polarized spin is injected from one FM contact, and then the current is modulated by modifying spin precession via Rashba effect (The Rashba effect is a momentum dependent splitting of spin bands in 2-D condensed matter systems like heterostructures and surface states. The splitting takes place due to the combined effect of atomic spin-orbit coupling and asymmetry of the potential in the direction perpendicular to the 2-D plane). Very fast, very dense memory and logic devices at extremely low power are expected to be developed soon. The possible revolutionary advances that are likely to take place are (i) Spin Quantum Devices (Spin FETs, LEDs, RTDs), (ii) Quantum Computing at Room T, and (iii) Complete Computer on a Chip.

Spin injection into semiconductors have been studied by Aronov *et al*. [18], who have reported that the spin injection *i.e.* the spin polarized current results from the passage of a current through a contact between a ferromagnet and a semiconductor. It has been pointed out that depending on the type of contacts; either the majority or the minority carriers are polarized. A detailed analysis of the influence of a magnetic field on such spin injection and conditions for its observation has been presented. Various other important studies have also been reported in the literature. Chopra and Maini [19] have emphasized the role of thin films for applications in spintronic devices. Chopra [20, 21] has discussed the Magnetic Tunneling Junctions, their Types, Structures and Significane, and also the New Materials and their Selection for Designing and Fabricating the Spintronic Devices. Ohno [22] has studied the III1-xMnxV Random Alloys (InMnAs and GaMnAs). Focus has been mainly on the III-Mn-V Semiconductors and their Heterostructures (GaMnAs, GaMnSb, InMnAs). Recently, emphasis has also been laid on the Digital Alloys: GaSb/InAs with Mn. So far, systematic study of digital GaAs: Mn alloys – Mn layer of thickness < 0.5 monolayer, with good structural properties (as confirmed by TEM, and X-ray Techniques) has been done. The variation of maximum TC with Mn layer, clustering of spin-spin coupling, phases, Anomalous Hall Effect., Magnetoresistance (initially positive MR followed by large negative MR in FM regime), and Thermally activated (hopping) conductivity in all samples have been studied and well understood. Some of the important results available in the literature can be summarized as: The III (1-x)MnxVs without precipitates have been grown below 300°C. However, it has been found that they are having poor optical quality. It has to be noted that mainly Mn is dopant (acceptor) in III-Vs; and it is generally not desirable for alloying. It has been reported that at low concentration (< 1%), they are insulating, and not FM. At higher Mn concentration (1% - 7%), they are metallic, and FM. Also, clustering of a few Mn ions takes place, though these are not precipitates. At even higher Mn concentration (> 8%), they are insulating, and not FM.

Also, in case of the MBE-grown GaMnAs Random Alloys, it has been reported that it is Ferromagnetic from about 3% - 7% Mn, and large density of holes comes with the territory, as Mn on Ga sites is an acceptor. The highest TC reported so far is - 110 K. Another interesting observation made is that the Mn++ - Mn++ interaction mediated by holes depends on the Mn and carrier (hole) densities. In case of the III-V/Mn digital alloys, it has been found that (i) The average Mn concentration increases and structural quality is improved, (ii) 2-D spin systems/carriers confined increase the TC, and (iii) The optical and transport properties are improved.

As is well known, the electric charge in bulk semiconductors and their heterostructures is manipulated, which forms the basis of the most of the modern electronic and optoelectronic devices. With the advent of the spin-dependent phenomena in semiconductors, emphasis has shifted to the technologies harnessing the spin of the electron in semiconductor devices. The added advantage in this case is that the spin degree of freedom has potential applications for fundamentally new functionality in the quantum domain ranging from storage to computation. This is precisely the reason, that it is playing at present a crucial role in the information technologies. Electronic processes in organic crystals and polymers have been described in great detail [23]. Lee *et al*. [24] have studied the 'Tuning of the Hyperfine Fields in Conjugated Polymers for Coherent Organic Spintronics', which are recently gaining a lot of attention because of their importance in the various applications. It has been discussed that in case of the organic spintronics, it is possible to have a direct coherent control of the spin population by means of pulsed electron spin resonance technique. In contrast to the previous work, which has focused on the electrical detection of coherent spin dynamics, this study demonstrates the equivalence of an all-optical approach, which allows to explore the influence of materials chemistry on the spin dynamics. It has been shown that deuteration of the conjugated polymer side groups weakens the local hyperfine fields experienced by the electron hole pairs, resulting in the lowering of the threshold for the resonant radiation intensity at which the coherent coupling and spin beating take place. It has to be emphasized that this technique offers a route to quantifying and tuning hyperfine fields in organic semiconductors. Rybicki *et al*. [25] have pointed out that the X rays produced during electron-beam deposition of metallic electrodes can affect the performance of organic spintronic devices, because the X rays generate traps of an activation energy of ~ 0.5eV in the organic used for such work. These traps in turn result in a substantial decrease in the spin-diffusion length in organic spin valves. However, for the case of the organic magnetoresistive (OMAR) devices, these traps enhance the magnetoresistance. Interestingly, the OMAR is an intrinsic magnetotransport phenomenon, which does not rely on spin injection. Attention has recently been paid to the Optimum Experimental Design for Extended Gaussian Disorder Modeled (EGDM) Organic Semiconductor Devices [26], who have presented an extended Gummel method to decouple the corresponding system of equations and used automatic differentiation to get derivatives with the required accuracy for the optimum experimental design (OED). They have considered two different cases, and shown that the linearized confidence regions of the parameters can be reduced considerably by applying the OED, resulting in new experiments with a different setup. Recently, unexpected Interaction between Organic Semiconductors [27] has also been observed, which is considered to be due to the strong bond between organic layers. These strange structures are still to be understood fully by the scientists. However, it is expected that these structures will form the basis for the novel electronic components made from organic semiconductors, which are now increasingly being used in smart phones and TVs. The recent findings are being exploited for developing new organic semiconductors. However, there is still a lot of more work to be done in this direction for establishing the industrial manufacturing processes, due to the requirements of the reproducibility and precision. Thus, it is safely concluded that the organic semiconductors are soon going to revolutionize the production of the devices with multiple functions capabilities, and also take the performance to a higher level of efficiency.

ACKNOWLEDGEMENTS

The author is grateful to the Dr. Nand Kishore Garg, Chairman, Maharaja Agrasen Institute of Technology, GGSIP University, Delhi for providing the facilities for carrying out this research work, and also for his moral support. The author is thankful to Dr. M. L. Goyal, Director, for encouragement. Thanks are also due to Dr. V. K. Jain, Deputy Director, for his support during the course of the work. The author is also grateful to Prof. V. K. Tripathi, Department of Physics, Indian Institute of Technology, Delhi for showing interest in the work.

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