Title: SPIN SEEBECK THERMOELECTRIC DEVICE AND ITS USES

Abstract: The present invention relates to a thermoelectric device which comprises: a layer of a non-magnetic material (NM) disposed over a layer of a magnetic material (F), wherein said layers form a first bi-layer junction (NM1/F1) of materials having spin Seebeck effect properties; and at least one second bi-layer junction of non-magnetic and magnetic materials (NM2/F2) having spin current transmission properties; wherein the second bi-layer junction (NM2/F2) is arranged to form, together with the first bi-layer junction (NM1/F1), a multilayer structure of materials having an amplified spin Seebeck effect compared to that of the first bi-layer junction (NM1/F1) alone. An optimized device can be obtained by stacking sequences of these bi-layers in a multilayered structure n(NM/F). The invention provides improved spin Seebeck thermoelectric devices, through a novel arrangement of materials which provide a substantial amplification of the spin pumped currents within the multilayer structure, thus generating enhanced voltage signals compared to those present in the prior art.

Published: with international search report (Art. 21(3))
FIELD OF THE INVENTION

The present invention belongs to the field of thermoelectricity and spin caloritronics. More specifically, the invention relates to thermoelectric devices based on the spin Seebeck effect.

BACKGROUND OF THE INVENTION

Currently, the environmental impact of climate change induced by greenhouse gas emissions manifests the importance of finding alternative energy harvesting technologies. In this context, thermoelectric energy conversion is a promising approach, since it allows the production of heating and cooling devices which are scalable and can be implemented in microelectronic technologies.

The field of thermoelectric phenomena applies the interaction properties of heat and electric currents for power generation and cooling systems, mainly through solid state devices that can be used for wasted heat energy recovery and cooling applications. These devices often rely on the known Seebeck and Peltier effects, which refer to the generation of an electric voltage by applying temperature gradients, and vice versa. Currently the most common uses of the thermoelectric effect relate to the development of thermometers that measure temperature differences with thermocouples, through Seebeck effect (junction of two different metals, selected according to the type of application and temperature range to be measured). Peltier modules for controlling the flow of heat for cooling installations are also known within the state of the art. Additionally, the use of the thermoelectric effect to generate electricity from heat energy is an emerging technical field that is in intense development.

However, the efficiency of thermoelectric devices is limited by a lower bound in the thermal conductivity, imposed by the mobile carriers through the Wiedemann-Franz law, since thermal conduction has to be minimized at the same time that electrical conduction has to be maximized. Common approaches to solve this problem focus on the disruption of the phonon conduction without affecting electronic carrier transport, by using the “phonon-glass electron-crystal” approach.
One possible candidate to overcome these limitations and improve the thermoelectric power conversion techniques lies on the recently discovered spin Seebeck effect (SSE), which involves the thermal generation of spin currents in a magnetic material (F) which, when injected into an attached non-magnetic material (NM) with high spin-orbit interaction, are converted into an electric voltage by means of the inverse spin-Hall effect (ISHE). The SSE has been experimentally established and observed in ferromagnetic metals, semiconductors and insulators, largely contributing to the expansion of the field of spin caloritronics, which is focused on the study of the correlation between charge, heat and spin currents in magnetic materials.

Theoretically, the spin Seebeck effect is understood as a result of the thermal non-equilibrium between magnons in the (F) material and conduction-electron spin accumulation in the (NM) material, which interact through the s-d exchange coupling $J_{sd}$ at the (F/NM) junction interface. Using a linear-response approach, the spin current injected into N is given by the following expression:

$$J_S = -G_S k_B (T_F^* - T_N^*),$$

where $T_F^*$ and $T_N^*$ are, respectively, the effective magnon temperature in the (F) material and the effective conduction electron temperature in the (NM) material. $G_S$ is a proportionality factor that depends on the size of the localized spin in the (F) material, the paramagnetic susceptibility and the spin-flip relaxation time in the (NM) material.

Increasing the efficiency of conventional thermoelectric devices remains still a challenge in the art, due to the need of optimizing conflicting properties of the employed materials. For instance, one needs to reduce the thermal conductivity ($\kappa$) of the devices, while also maximizing their efficiency, measured by the effectiveness parameter $Z$ (or figure of merit), represented by the expression $Z=S^2\sigma/\kappa$, where $S$ is the Seebeck coefficient and $\sigma$ is the electrical conductivity. Thus, in order to increase $Z$, it is necessary to increase the product $S^2\sigma$, while reducing $\kappa$. However, this requirement conflicts with the behaviour of $\sigma$ and $\kappa$, due to the intrinsic proportionality of these two quantities for the same given material. Therefore, using two materials with different properties in the SSE devices provides improved design for increased performance, compared to conventional thermoelectric devices. Thus, the magnetic material (bad metal or insulator) can be optimized for a maximum thermal gradient, with minimum application of energy, while maximizing the spin current conversion of the material used to obtain by ISHE the electric potential, thus obtaining the decoupling of the two parameters.
The observation of the SSE in magnetic insulators is of great interest for potential applications in thermoelectric power generation devices, since it allows the generation of electric voltages without the Joule heating associated to mobile charge carriers. This advantage offers the possibility of exploring potential applications of magnetic oxide materials to thermoelectric devices, which are not generally suitable for conventional thermoelectrics (for example, in Seebeck- and Peltier-effect applications). Another advantage is that SSE involves the properties of at least two different materials that can be optimized independently. However, the main roadblock for the application of the spin Seebeck effect to thermoelectric devices is the low value of the obtained voltage signals, typically of the order of 100 nV/K.

Due to the above mentioned considerations, finding mechanisms for enhancing the output signals is of utter importance and can have a huge impact on the application for thermoelectric devices. Consequently, a substantial improvement of the observed signal (voltage) in these devices is still needed for realistic industrial thermoelectric applications. In this sense, although there are approaches aimed at the enhancement of the output power by using a plurality of thermoelectric devices connected in series (forming a thermopile structure), the small signal of each individual device for these systems remains yet a strong limitation in the field. The invention described herein is intended to solve this technical problem, through a thermoelectric device and thermoelectric generation method which apply the SSE concept to a novel arrangement of materials, providing a substantial amplification of the spin pumped currents within the device, thus generating enhanced voltage signals compared to those present in the prior art devices.

**BRIEF DESCRIPTION OF THE INVENTION**

An object of the invention is thus obtaining novel thermoelectric devices with enhanced output voltage signals compared to the SSE devices known in the art. To achieve this object, the embodiments of the invention are related to the amplification of the electric potential generated by the thermoelectric conversion due to the spin Seebeck effect, converting temperature differences in electric potential differences in a material by means of spin currents. In the conventional Seebeck effect the thermoelectric generation is obtained by means of temperature gradients in conductive and semiconductive materials. The SSE, measured up to now in (ferro/ferri)magnetic (F) and non-magnetic
conductor (NM) bi-layers, is based on the generation of spin currents resulting from the application of thermal gradients on magnetic materials. Thus, a generated spin current becomes an electric potential difference when said current is injected into an associated non-magnetic material having a high spin-orbit interaction, by means of the inverse spin-Hall effect.

In a preferred embodiment of the invention, the thermoelectric device comprises:
- a layer of a non-magnetic material (NM1) disposed over a layer of a magnetic material (F1), wherein said layers form a first bi-layer junction (NM1/F1) of materials having spin Seebeck effect properties;
- and at least one second bi-layer junction of non-magnetic and magnetic materials (NM2/F2) having spin current transmission properties;

wherein the second bi-layer junction (NM2/F2) is arranged to form, together with the first bi-layer junction (NM1/F1), a multilayer structure of materials having an amplified spin Seebeck effect compared to that of the first bi-layer junction (NM1/F1) alone.

The SSE amplification achieved through the present invention is thus a consequence of the arrangement of several bi-layer junctions in contact forming a multilayer structure, wherein the interfaces of the bi-layer pairs provides an increased voltage which depends on the number of bi-layers in the multilayer structure.

In another preferred embodiment of the invention, at least a second bi-layer junction (NM2/F2) of non-magnetic and magnetic materials in the device has also spin Seebeck effect properties.

In a further preferred embodiment of the invention, the thickness of the magnetic material layers is about the characteristic magnon diffusion length of said materials (40 – 100 nm) and the non-magnetic material layers of the order or thinner than the characteristic length for spin diffusion of said materials (3 – 100 nm).

Another object of the invention relates to a thermopile comprising a plurality of thermoelectric devices according to any of the embodiments described in the present document, wherein said devices present an amplified SSE and are connected in series and/or in parallel by electrical connections.

In a preferred embodiment of the invention, the devices forming the thermopile are
arranged in parallel and apply connections in a zigzag-type structure. More preferably, the thermopile comprises a plurality of zigzag-type structures electrically connected in parallel. In alternative embodiments of the invention, the connections comprise an electrically conductive material with low spin-orbit coupling and no spin Seebeck effect properties (for example, Cu).

Another object of the invention relates to a method for generating an amplified spin-Seebeck-effect electric potential, which comprises the steps of providing a plurality of bi-layer junctions forming a thermoelectric device according to any of the embodiments described in this document; the plurality of NM1/F1 interfaces changes the boundary conditions and provides continuity for the propagation of the spin current, resulting in an amplified spin current in the multilayer structure, with maximum values at the central layers. Preferably, the electric potential is generated in the region corresponding to the central bi-layer junction (NM1/F1) of the multilayer structure of the device.

A further object of the invention relates to the use of a device according to any of the embodiments described in this document, for amplifying the electric potential generated in said device.

As for the industrial applications of the present invention, they preferably apply to the field of energy conversion, and particularly to the development of means for converting heat sources in electric power sources. Other potential uses of the invention relate to the development of temperature sensors (i.e. biosensors) or heat flow controllers.

Finally, the main advantage of the proposed invention relates to the amplification of SSE materials in multilayer structures. This represents an advance in the known applications of this effect, implying further technical advantages over conventional thermoelectric materials. In this sense, the invention allows the generation of spin currents which do not dissipate energy. The invention also allows the decoupling of the parameters governing the thermoelectric performance, allowing its optimization for its different embodiments, based on a different principle, the SSE.

In addition to the above, the invention has the advantage that it can be carried out using conventional techniques of forming thin films and layers of materials, as for example physical vapor deposition, which are widely used in industry. Other known techniques such as optical lithography techniques may also be relevant in manufacturing
applications of the invention, mainly in the thermopiles.

DESCRIPTION OF THE FIGURES

Fig. 1 describes a thermoelectric device according to a preferred embodiment of the present invention. The proposed multilayer device is compared to the usual NM/F bi-layer.

Fig. 2 illustrates the generation of spin currents in a thermoelectric device according to a preferred embodiment of the present invention. The spin magnon current is thermally generated and converted into an electron spin current in the non-magnetic material (NM) at the interfaces. This results in a maximum spin current in the middle of the multilayer.

Fig. 3 shows alternative multilayer arrangements according to two different embodiments of the present invention. This configuration profits from the maximum voltage generated in the middle of the multilayer, according to the theoretically predicted maximum of the spin current. Figure 3(a) shows an embodiment in which all non-magnetic conducting layers have a high spin-orbit coupling (NM(SO)). Figure 3(b) shows a multilayer arrangement in which only the central NM(SO) layer presents high spin-orbit coupling, while the other non-magnetic conducting layers (NM) present negligible spin-orbit interaction. "FI" in the figure refers to a ferromagnetic insulating material (typically a ferromagnetic oxide).

Fig. 4 shows two schemes of thermopile systems based on the connection of SSE thermoelectric devices according to a preferred embodiment of the present invention. In Figure 4(a), the SSE devices are arranged in parallel and apply connections in a zigzag-type structure. Figure 4(b) shows a plurality of zigzag-type structures electrically connected in parallel.

Fig. 5 shows the spin Seebeck effect of a single Pt/Fe$_3$O$_4$ bi-layer. Fig. 5(a) is a schematic illustration of the measurement geometry. Fig 5(b) shows the results obtained for a single Pt/Fe$_3$O$_4$ bi-layer, for different applied thermal gradients at 300 K. Fig 5(c) shows the dependence of the magnetic saturation $(V_{sat})$ measured at 7 kOe for the different magnitudes of the applied temperature difference $(\Delta T)$ across the sample (the line shows the fit to a linear dependence).
**Fig. 6** shows the results of a spin Seebeck multilayer system according to the invention. Fig. 6(a) depicts the magnetic field dependence of the spin Seebeck effect measured for the different multilayer samples at 300 K. Fig. 6(b) shows the dependence of $V_{\text{sat}}^y$ measured at 7 kOe as a function of the number of Pt/Fe$_3$O$_4$ bi-layers (n).

**Fig. 7** shows a comparison between the spin Seebeck effect response at 300 K for $1 \times (\text{Pt} (t_h)/\text{Fe}_3\text{O}_4 (t_f))$, $3 \times (\text{Pt} (t_h)/\text{Fe}_3\text{O}_4 (t_f))$ and $\text{Pt} (t_h)/\text{Fe}_3\text{O}_4 (t_f)/2 \times (\text{MgO}(8\text{nm})/\text{Fe}_3\text{O}_4 (t_f))$ bi-layer arrangements; $t_N=7$ and $t_f=23$ nm.

**Fig. 8** illustrates the dependence of the spin Seebeck effect response of a multilayer sample on the parameters of the NM layers. Figure 8(a) shows the SSE for a single $\text{Pt} (t_h)/\text{Fe}_3\text{O}_4 (t_f)$ bi-layer and a $6 \times (\text{Pt} (t_h)/\text{Fe}_3\text{O}_4 (t_f))$ multilayer measured at 300 K, where $t_h=7$ and $t_f=23$ nm. Figure 8(b) shows the SSE for a $6 \times (\text{Pt} (t_h)/\text{Fe}_3\text{O}_4)$ with Pt thicknesses of 7 and 17 nm.

**DETAILED DESCRIPTION OF THE INVENTION**

Now we proceed to describe some of the preferred and not limiting embodiments of the invention, in relation to the numerical references of the Figures 1-8 herein.

As discussed in previous sections, the present invention provides means by which the limitations of the known SSE devices are overcome by the use of material arrangements, based on a sequence of layers forming a non-magnetic material (NM1) interspersed with layers of a magnetic material (F) (Figure 1(b)). More specifically, the thermoelectric device of the invention is partially based on the material structures forming a typical SSE device, comprising a layer of a non-magnetic material (NM1) disposed over a layer of magnetic material (F1), wherein said layers form a first bi-layer junction (NM1/F1) of materials having spin Seebeck effect properties. Additionally, the device of the invention comprises at least one second bi-layer junction of non-magnetic and magnetic materials (NM2/F2), having spin current transmission properties.

The ferromagnetic/ferrimagnetic material (F) might be any oxide containing Fe, Co or Ni. For instance, it can be used: Fe$_3$O$_4$, $\gamma$-Fe$_2$O$_3$, CoFe$_2$O$_4$, NiFe$_2$O$_4$, (Ni, Zn)Fe$_2$O$_4$, or other ferrimagnetic spinel ferrites with the formula AB$_2$O$_4$, where A and B represent metal cations, with B being usually Fe. Hexagonal ferrites: BaFe$_{12}$O$_{19}$, SrFe$_{12}$O$_{19}$, Yttrium Iron
and other Garnet oxides: $\text{Y}_3\text{Fe}_5\text{O}_{12}$, $\text{Y}_3\text{Fe}_5\text{Co}_{0.5}\text{Ga}_{0.5}\text{O}_{12}$. Magnetic oxides: perovskites and double perovskites.

As possible non-magnetic materials (NM) for the first bi-layer junction (NM1/F1) we can consider: Pt, Au, Pd, Bi, W, Ta, Mo, Nb or other conducting materials with large spin-orbit coupling.

Contrary to the known SSE devices, the second bi-layer junction (NM2/F2) of the present invention is arranged to form, together with the first bi-layer junction (NM1/F1), a multilayer structure of materials having an amplified spin Seebeck effect compared to that of the first bi-layer junction (NM1/F1) alone. This amplification is mainly achieved from the amplification of the generated spin current though the arrangement of the bi-layer junctions (NM/F) as a multilayer structure. In fact, as shown in Figure 2, when a temperature gradient on a magnetic layer is applied, a spin/magnon current (Fig. 2(a)) is generated. On the other hand, when a plurality of n×(NM/F) layers is arranged, the heat generated spin current is amplified (especially in the layers corresponding to the central core), as a result of the boundary conditions of the structure (Fig. 2(b)).

Due to said amplification in the central region of the multi-layer arrangement, it is thus possible to carry out alternative embodiments of the invention in which high spin-orbit-coupling materials are used in the layers corresponding only to the region of maximum amplification, while the materials used in other layers are only required to transmit the spin, but not necessarily to convert it (for example, Cu). As possible spin transmitting materials with no spin current conversion we can consider: Cu, Ag, Al, or other non-magnetic materials with low spin-orbit coupling. Spin to current converting materials can include: Pt, Au, Pd, Bi, W, Ta, Mo, Nb or other materials with high spin-orbit coupling. As shown in Figure 3, it is therefore possible to design embodiments of the invention where, even employing materials with high spin-orbit coupling only in the central regions (such as Pt in the Figure), the obtained amplification effect would be similar to that obtained using these materials in all layers.

Complementary to other embodiments of the invention, for a further increase of the generated electric potential, the present invention also proposes manufacturing a thermopile according, for example, to any of the schemes depicted in Figure 4. The thermopile preferably comprises a plurality of thermoelectric devices, each of them presenting an amplified SSE, wherein said devices are connected in series and/or in
parallel by electrical connections. This provides means of increasing the output voltage and the electric power of a thermoelectric system. In Figure 4(a), the SSE devices are arranged in parallel and apply connections in a zigzag-type structure. Figure 4(b) shows a plurality of zigzag-type structures electrically connected in parallel.

By using optical lithography techniques, it is possible to create a pattern of parallel wires (the pattern can be transferred to the upper layer or the entire NM multilayer structure), where these wires must be electrically interconnected with a material forming a zigzag structure. This material is not directly related to the SSE itself, but acts as an electrical connection between thermoelectrically active elements. The additive effect of potential increase within thermopile structures depends on the density of the pattern area of parallel wires, although further increases proportional to the number of threads would also occur in the electric signal.

Description of a preferred embodiment of the invention:

As a non-limiting example of the invention, a device based on the spin Seebeck effect of magnetic multilayers is hereby described. More specifically, the device comprises an arrangement of magnetic multilayers formed by repetition of platinum/magnetite (Pt/Fe$_3$O$_4$) bi-layer junctions grown on MgO (001) substrates. Magnetite is an oxide with a characteristic metal-insulator transition, $T_v \sim 125$K (Verwey transition). It also presents very interesting magnetic properties, being a half-metal with full spin polarization and a high Curie temperature. These properties make it an interesting material for spintronic studies, since it provides a robust source of spin.

For this embodiment of the invention, the spin Seebeck effect of a multilayer $n$×(Pt/Fe$_3$O$_4$) device is studied, by measuring the thermal voltage generated on the topmost Pt layer only. As it will be shown, an enhancement of the measured voltage with the number of bi-layers ($n$) is observed in the device. Contrary to other thermoelectric devices, the observed increase of the spin Seebeck effect is not due to a modulation of the thermal conductivity due to superlattice structure, but is of intrinsic magnetic origin, as a consequence of the incremental effect of the thermal spin injection across the multiple Pt-F$_3$O$_4$ interfaces in the multilayer device.

The multilayers were fabricated by repeated growth of Pt($t_h$)/Fe$_3$O$_4$($t_f$) bi-layers on MgO(001) substrates, where $t_f=34$ nm and $t_h=17$ nm. The samples have rectangular
shape with dimensions $L_y=7$ mm, $L_x=2$ mm and $L_z=0.5$ mm. The spin Seebeck effect voltage was monitored on the topmost Pt layer only. Figure 5(a) shows the geometry used for the longitudinal spin Seebeck effect measurements, a thermal gradient ($\nabla T$) is applied in the $z$ direction and the generated spin current is converted to an electric voltage ($V_y$) in the Pt film ($y$ direction), that is monitored while a sweeping magnetic field is applied (x direction). The SSE voltage, $V_y$, for the single Pt/Fe$_3$O$_4$ bi-layer system is shown in Figure 5(b) for several applied temperature differences between the top and bottom of the sample ($\Delta T$). The observed signal scales linearly with the applied thermal gradient as shown in Fig. 5(c), which shows the voltage measured at magnetic saturation ($V_{sat,y}=V_y(7 \text{ kOe})$), the solid line shows the fit to a linear dependence.

From the slope of this curve and considering the sample geometry, we can extract the spin Seebeck coefficient, which is given by the following expression: $S_{SSSE}=(V_{sat,y}/\Delta T)(L_y/L_x)$. It is known that despite Fe$_3$O$_4$ being electrically conductive above $T_N$, the response observed in the Pt/Fe$_3$O$_4$ device is highly dominated by the SSE. This is due to the resistance mismatch between Pt and Fe$_3$O$_4$ layers, being Fe$_3$O$_4$ two orders of magnitude more resistive than Pt. The contribution from the anomalous Nernst effect (ANE) of magnetite to the measured signal is estimated to account for less than 1% of the observed signal in the measurements, therefore we can infer that the observed signal is mainly due to the spin Seebeck effect of Pt/Fe$_3$O$_4$.

Spin Seebeck effect measurements were performed on $n$x(Pt/Fe$_3$O$_4$) samples by probing the spin Seebeck voltage generated on the topmost Pt layer only. The results obtained at room temperature as a function of the number of bi-layers, $n$, are shown in Figure 6(a). Surprisingly, the observed voltage scales with the number of Pt/Fe$_3$O$_4$ bi-layers. Figure 6(b) shows the spin Seebeck voltage measured at magnetic saturation ($V_{sat,y}$), where it can be clearly seen that the observed voltage scales with the number of Pt/Fe$_3$O$_4$ bi-layers in the system. Since this voltage is directly proportional to the injected spin current one would not expect such an enhancement if the thermal gradient is constant across the sample. Thermoreflectance measurements show only a weak dependence of the thermal conductivity upon increasing the number of bi-layers, obtaining an average value of 2.9±0.2 W/mK for all the samples measured, thus discarding the reduction of the thermal conductivity in the multilayer system as the origin of our observations.

Among the remaining possibilities we can consider; a difference in the boundary conditions for the magnon and/or electron subsystems in the different interfaces, which
could create a modulation of the difference between the magnon temperature in the ferromagnet (T_F) and the phonon temperature in the non-magnetic layer (T_N), with the temperature difference between these two quantities increasing with a higher number of interfaces. Another possibility is an incremental effect of spin pumping across the multiple Pt-Fe₃O₄ interfaces of the multilayer system. In this case, the overall result would be an amplified spin current which will translate in a higher measured voltage through the inverse spin Hall effect (ISHE).

We also describe the results of the performed measurements on a multilayer device in which the inner Pt (17 nm) interlayers of the samples were substituted by MgO interlayers, leaving only the topmost Pt layer. These structures do not disrupt the thermal transport across the multilayer structure, but the spin current vanishes across the MgO spacer. Figure 7 shows the comparison between the measurements performed on the 1x(Pt/Fe₃O₄), 3x(Pt/Fe₃O₄) and Pt/Fe₃O₄/2x(MgO/Fe₃O₄) sample devices at 300 K. It is interesting to observe that, in the multilayer system with MgO interlayers, the signal is strongly reduced. MgO is an insulator and the heat is carried by the phonons in this system, therefore heat transport is not disrupted while the electron/spin transport across the MgO spacer layer is negligible. This result is a clear indication that the observed voltage enhancement is not a consequence of a variation of the thermal boundary conditions in our multilayer system, but points to a purely spin current effect. We can see that the value of V_y at magnetic saturation for the Pt/Fe₃O₄/2x(MgO/Fe₃O₄) multilayer sample is comparable to the one obtained for a single Pt/Fe₃O₄ sample, pointing out to the importance of the number of the Pt-Fe₃O₄ interfaces to the observed effect.

The role of the Pt interlayers thickness in the observed effect was studied by measuring the SSE of a 6x(Pt/Fe₃O₄) multilayer, with Pt layers of a thickness of 7 nm, of the order of the Pt spin diffusion length. Fig. 8(a) shows the results of the 6x(Pt/Fe3O4) multilayer sample with a SSE of 25 μV/K, a 5-fold increase in magnitude compared to the one observed for a single bi-layer of same thickness parameters (5 μV/K). Fig. 8(b) shows the comparison between a 6x(Pt(t₀)/Fe3O4) with t₀ = 7 and 17 nm, where the increased signal in the case of the multilayer structure with 7 nm Pt interlayers is clearly observed, clearly indicating that the effect is related to the spin current propagation across the Pt interlayers.
For the present embodiment of the invention, the observed results can be qualitatively explained considering a bulk magnon spin current in the F layer. We impose the following boundary conditions: a) the spin/magnon current should vanish at the top/bottom surface of the multilayer structure, b) the continuity of the electron/magnon spin currents at the Pt/Fe₃O₄ interfaces. Under these conditions, an amplification of the spin current in the multilayer system is predicted, obtaining a maximum spin current value at the central interlayers of the structure (see Fig. 2b), thus resulting in an enhanced SSE.

The observed enhancement of the spin Seebeck effect provides a new mechanism for the amplification of the thermally generated voltage, the multiple Pt-Fe₃O₄ interface changes the length scale and boundary values for the spin current in the Pt interlayers, with maximum spin current in the central Pt layers. We observe an enhancement of one order of magnitude of the SSE, obtaining a thermal voltage of 25μV/K. This result opens a promising route of development for potential application of the spin Seebeck effect on thermoelectric devices.
CLAIMS

1.- Thermoelectric device comprising:

a layer of a non-magnetic material (NM1) disposed over a layer of a magnetic material (F1), wherein said layers form a first bi-layer junction (NM1/F1) of materials having spin Seebeck effect properties;

and at least one second bi-layer junction of non-magnetic and magnetic materials (NM2/F2) having spin current transmission properties;

said device being characterized in that the second bi-layer junction (NM2/F2) is arranged to form, together with the first bi-layer junction (NM1/F1), a multilayer structure of materials having an amplified spin Seebeck effect compared to that of the first bi-layer junction (NM1/F1) alone.

2.- Device according to the preceding claim, wherein at least a second bi-layer junction (NM2/F2) of non-magnetic and magnetic materials has also spin Seebeck effect properties.

3.- Device according to any of the preceding claims, wherein the magnetic material (F1,F2) comprises an oxide containing Fe, Co or Ni.

4.- Device according to the preceding claim, wherein the magnetic material (F1,F2) comprises one or more of the following: Fe$_3$O$_4$, γ-Fe$_2$O$_3$, CoFe$_2$O$_4$, NiFe$_2$O$_4$, (Ni, Zn)Fe$_2$O$_4$, a ferrimagnetic spinel ferrite with the formula AB$_2$O$_4$, where A and B represent metal cations, with B being Fe, a hexagonal ferrite, a garnet oxide, a magnetic oxide.

5.- Device according to any of the preceding claims, wherein the non-magnetic material (NM1, NM2) comprises one or more of the following: Pt, Au, Pd, Bi, W, Ta, Mo, Nb.

6.- Device according to claim 1, wherein the non-magnetic material (NM2) of the second bi-layer junction (NM2/F2) comprises one or more of the following: Cu, Ag, Al.

7.- A device according to any of the preceding claims, wherein the thickness of the non-magnetic/magnetic material (NM/F) layers is equal to or less than the characteristic spin/magnon diffusion length of said materials.
8. A device according to the preceding claim wherein the thickness of the non-magnetic layers is between 3-100 nm and the thickness of the magnetic layers is between 40-100 nm.

9. Thermopile comprising a plurality of devices according to any of claims 1-8, wherein said devices are connected in series and/or in parallel by electrical connections.

10. Thermopile according to the preceding claim, wherein the devices are arranged in parallel and comprise connections applied in a zigzag-type structure.

11. Thermopile according to the preceding claim, comprising a plurality of zigzag-type structures electrically connected in parallel.

12. Thermopile according to any of claims 9-11, wherein the electrical connections comprise electrically conductive material with no spin Seebeck effect properties.

13. Method for generating an amplified spin-Seebeck-effect electric potential, characterized by providing a plurality of bi-layer junctions forming a device according to any of claims 1-8, and generating a spin current through the bi-layer junctions (NM1/F1) of said device.

14. Method according to the preceding claim, wherein the electric potential is generated in the region corresponding to the central bi-layer junction (NM1/F1) of the multilayer structure of the device.

15. Use of a device according to any of claims 1-8 for amplifying the electric potential generated in a bi-layer junction (NM1/F1) of said device.
FIG. 1

a. Single bilayer

b. Multilayer

- Non-magnetic layer (NM)
- Ferromagnetic layer (F)

Heat source
Reduce output impedance

FIG. 4

- multilayer
- electrical contact
FIG. 5
**FIG. 6**

(a) Plot of $V_{ML} / \Delta T$ (µV/K) vs. $H$ (kOe) for different values of $n$: $n = 1$, $n = 2$, $n = 3$, $n = 6$. The temperature is $T = 300$ K.

(b) Graph showing $V_{ML}^{\text{sat}} / \Delta T$ (µV/K) vs. the number of $n \times (\text{Pt/Fe}_2\text{O}_4) / \text{MgO}(001)$ bilayers. The temperature is $T = 300$ K.
FIG. 7
FIG. 8
INTERNATIONAL SEARCH REPORT

A. CLASSIFICATION OF SUBJECT MATTER

INV. H01L37/00
ADD.

According to International Patent Classification (IPC) or to both national classification and IPC

B. FIELDS SEARCHED

Minimum documentation searched (classification system followed by classification symbols)
H01L

Documentation searched other than minimum documentation to the extent that such documents are included in the fields searched

Electronic data base consulted during the international search (name of data base and, where practicable, search terms used)
EPO-Internal, WPI Data

C. DOCUMENTS CONSIDERED TO BE RELEVANT

<table>
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<tr>
<th>Category</th>
<th>Citation of document, with indication, where appropriate, of the relevant passages</th>
<th>Relevant to claim No.</th>
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<td>X</td>
<td>WO 2013/047253 A1 (NEC CORP [JP]; ISHIDA MASAHIKO [JP]; KIRIHARA AKIHIRO [JP]; KOUUMOTO SH) 4 April 2013 (2013-04-04) paragraph [0031]; figure 6</td>
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