Hou et al. *NPG Asia Materials* (2019) 11:35
https://doi.org/10.1038/s41427-019-0135-9

NPG Asia Materials

PERSPECTIVE Open Access

Spin transport in antiferromagnetic insulators: progress and challenges

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Abstract

Spin transport is a key process in the operation of spin-based devices that has been the focus of spintronics research for the last two decades. Conductive materials, such as semiconductors and metals, in which the spin transport relies on electron diffusion, have been employed as the channels for spin transport in most studies. Due to the absence of conduction electrons, the potential to be a spin channel has long been neglected for insulators. However, since the demonstration of spin transmission through a ferromagnetic insulator, it was realized that insulators with magnetic ordering can also serve as channels for spin transport. Here, the recent progress of spin transport in antiferromagnetic insulators is briefly described with an introduction to the experimental techniques. The observations regarding the temperature dependence of spin transmission, spin current switching and the negative spin Hall magnetoresistance are discussed. We also include the challenges for developing the functionality of antiferromagnetic insulators as well as the unresolved problems from the experimental observations.

A core mission for spintronics is to develop spin-based devices in which information is processed by spin rather than charge[1]. Efficient spin transport is a prerequisite for the operation of spin-based devices. If spin transport can be further controlled by some external parameters, e.g., gate voltage, a spin transistor can be realized[2]. Following these visions from the pioneers, spin transport has been studied extensively in various materials, and different approaches have been developed for its manipulation[3].

It is likely that the mainstream materials for spin transport have been semiconductors and metals in which the spin current is essentially electron diffusion with a difference between the up-spin and down-spin electron chemical potentials because the spin current was initially introduced and demonstrated with conduction electrons[4,5]. However, since a spin current can generally be considered as a flow of angular momentum, electrons should not be the only choice for the carrier of spin. In principle, systems comprising any particle or quasiparticle with an angular momentum degree of freedom can host spin current. For instance, the spin transport mediated by magnons in $Y_3Fe_5O_{12}$ (YIG), a ferrimagnetic insulator, was demonstrated by Kajiwara et al.[6]. From then on, the study of spin transport in ferromagnetic insulators has been a topic of interest in the spintronics field.

Since the spin current in ferromagnetic insulators is believed to be mediated by magnons, antiferromagnetic insulators (AFMI), which also host magnons, hold potential for spin transport as well. Indeed, the demonstration of spin transport through antiferromagnetic insulators was first achieved by Wang et al.[7], which was followed by similar results from other groups[8,9]. This breakthrough included a new class of materials for spin transport, which opened up new possibilities for future device development.

Here, the experimental configurations for the study of spin transport in AFMI are briefly introduced with a discussion of the observation results. Figure 1 illustrates

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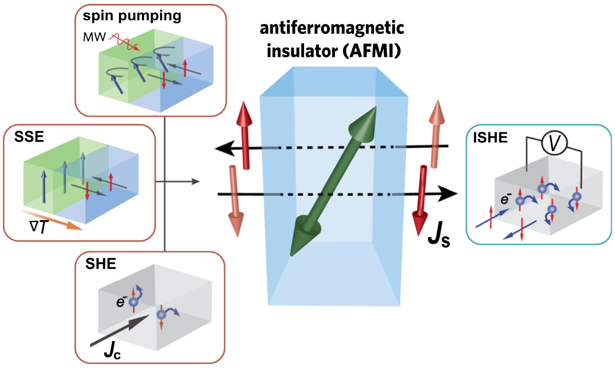

Fig. 1 Illustrations of spin transport in an antiferromagnetic insulator and various schemes of spin current generation and detection. A spin current can be injected into an antiferromagnetic insulator alternatively by the spin pumping process, spin Seebeck effect (SSE) or spin Hall effect (SHE). The transmitted spin current can be detected with a voltage signal via the inverse spin Hall effect

the essential processes for the observation of spin transport in AFMI from left to right: spin current generation, spin transmission, and spin current detection. The spin pumping process, spin Seebeck effect or spin Hall effect can be used to generate and inject a spin current into an AFMI, in which the driving forces are microwave, temperature gradient and charge current, respectively[10–12]. The spin current transmitted through an AFMI can be detected by a voltage in an adjacent heavy metal via the inverse spin Hall effect[10].

The sample structure used in the ref. [7], a Pt/NiO/YIG trilayer, is taken as an example and illustrated in Fig. 2a. A spin current is injected into NiO, which is an antiferromagnetic insulator (AFMI), from $Y_3Fe_5O_{12}$ by spin pumping and detected by the inverse spin Hall effect (ISHE) in Pt[10]. Surprisingly, the ISHE voltage in the Pt/NiO/YIG device is even larger than that in the Pt/YIG device when the NiO is approximately 1 nm, which means that the spin current is somehow enhanced by the presence of the NiO interlayer. Such a counterintuitive result was also found in the spin Seebeck measurement for Pt/NiO/YIG[13], which is shown in Fig. 2e. An instant question following these studies is: what is the optimal condition for spin transport in an AFMI? It is important both for the potential of AFMIs in further applications and for an understanding of the microscopic mechanism of spin transport. The temperature dependence of the damping constant in a permalloy/Cu/IrMn/Al device initially shed light on this issue, in which IrMn is an antiferromagnetic alloy. These results show that the spin current injection by spin pumping has an anomalous enhancement at a temperature that increases with the IrMn thickness, as shown in Fig. 2f[14]. Although it is quite challenging to determine the ordering temperature of the IrMn film in their devices, which is below 1.5 nm, it was argued that the spin pumping enhancement temperature may be close to the Néel temperature.

Although the IrMn alloy is metallic rather than insulating, the result undoubtedly highlights the intriguing behavior of spin transport in the phase transition regime of antiferromagnets.

Shortly after the publication of the spin pumping result for IrMn[14], spin transmission was reported in a Pt/CoO/YIG device by a temperature-dependent spin pumping inverse spin Hall measurement and directly determined the Néel point of the CoO by X-ray linear dichroism (XMLD) at the same time[15]. Figure 2b plots the representative ISHE voltage (V_{ISHE}) measured with a magnetic field that was swept at different temperatures, and Fig. 2c is the temperature dependence of the V_{ISHE}, which shows a peak at approximately 200 K. Figure 2d is the result of the XMLD measurement from the same structure, which yields a Néel temperature of 210 K with an error bar of 10 K. Thus, the spin current transmission in CoO does have an enhancement around the Néel temperature, which is consistent with the result in[14]. Similar temperature dependence was also observed by the spin Seebeck measurement in similar structures[13].

All of these results strongly suggest that the thermal magnon population is important for spin transport in AFMI. Nevertheless, since spin fluctuation cannot be neglected in the vicinity of the Néel temperature, it is still a theoretical issue whether spins are transported in the form of a magnon or a spin fluctuation. On the other hand, a key experiment that could determine whether the transmitted spin current is coherent is still not available[16]. The evanescent mode is argued to cause the amplification of spin current in NiO, but it seems hard to explain the vanishing spin transmission in AFMIs at low temperatures[7,17]. Further progress on the spin transport mechanism determination may be made by the temperature dependence of the AC ISHE[18,19], from which the coherence of the AC spin current can be checked.

So far, the studies mentioned above were more about the characterization of the spin transport property in AFMIs[7–9,13–15], while the control of the spin transport in the AFMIs, e.g., the switching of a spin current, which is indispensable for next-step applications, has not been achieved yet. Recently, isothermal switching of a spin current was demonstrated in Cr_2O_3, in which the spin transmission modulation is greater than 500% under a magnetic field[20]. Figure 3a shows the illustrations for the experimental configuration, and Fig. 3b is the cross-section TEM image of the device. Here, the YIG/Cr_2O_3/Pt structure is employed, in which a spin current is driven by a temperature gradient, ∇T, from the YIG into the Cr_2O_3 by the spin Seebeck effect[11,21,22]. In contrast to the samples used in a previous study, the Cr_2O_3 layer has a well-aligned out-of-plane Néel vector due to its single

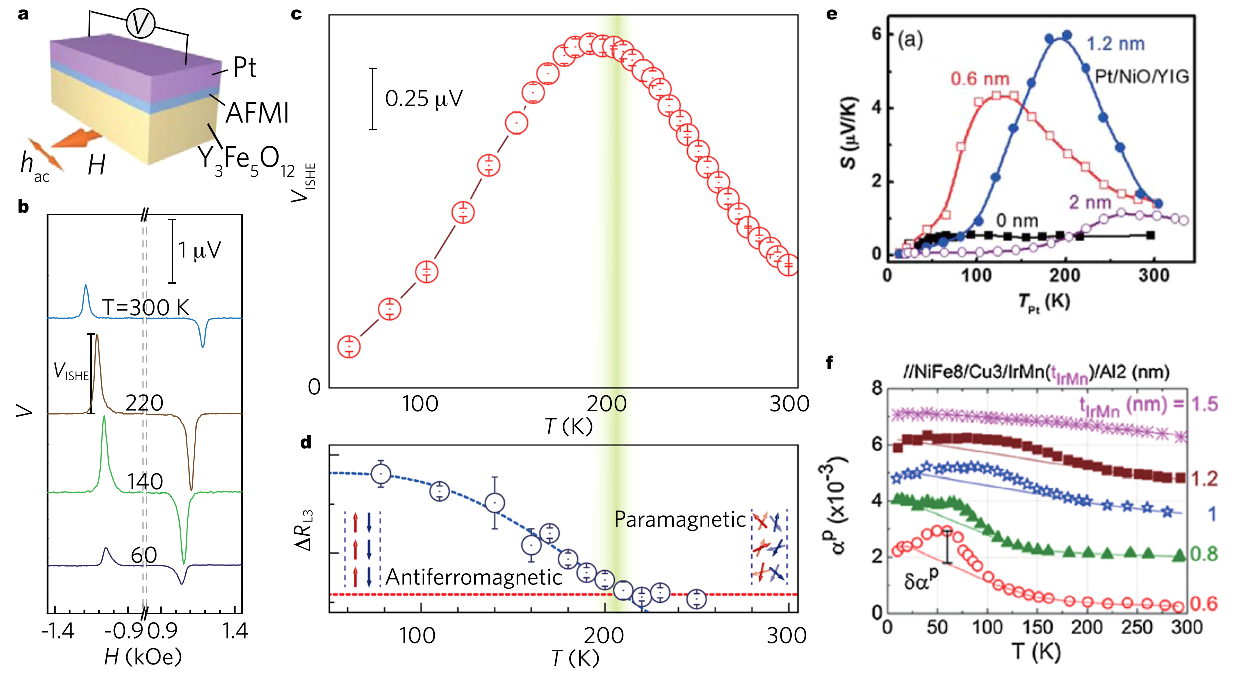

Fig. 2 a A schematic illustration of the device structure for spin transport in an antiferromagnetic insulator (AFMI), e.g., NiO or CoO. The ferrimagnetic insulator $Y_3Fe_5O_{12}$ serves as the spin injector, and the transmitted spin current is detected via the inverse spin Hall effect in Pt. **b** The inverse spin Hall voltage observed in a magnetic field scan at various temperatures in a Pt/CoO (6 nm)/YIG device. **c** Temperature dependence of the inverse spin Hall voltage in Pt/CoO (6 nm)/YIG. **d** Temperature dependence of the X-ray linear dichroism signal ΔR_{L3} in Pt/CoO (6 nm)/YIG, from which the Néel temperature is determined. **e** The spin Seebeck thermal power in a series of Pt/NiO/YIG samples with different NiO thicknesses. **f** The temperature dependence of the damping constant of permalloy/Cu/IrMn/Al samples after background subtraction for different IrMn thicknesses. **e** Adapted from the ref. [13]. **f** Adapted from the ref. [14]

crystalline structure and uniaxial magnetic anisotropy. Figure 3c plots the temperature dependence of the spin Seebeck voltage in a YIG/Cr_2O_3/Pt device measured with an in-plane magnetic field, which shows a sudden transition from the spin conductor state to the spin nonconductor state within 14 K of the Néel temperature. Such behavior is in sharp contrast to the gradual decay of spin transmission below the Néel temperature in CoO and NiO[13,15]. The suppression of spin transmission in the antiferromagnetic phase can be understood by the symmetry requirement of the magnon spin current: the spin polarization of magnons must be parallel to the Néel vector, which is different from the arbitrary spin polarization direction in electron systems. This property can be inferred from the spin transmission in the YIG, in which the spin current is blocked when the magnetization of YIG is perpendicular to the direction of the injected spin from Pt. Thus, the configuration in Fig. 3a corresponds to an "OFF" state for spin transmission due to the orthogonal relative orientation between the magnetization of YIG and the Néel vector of Cr_2O_3.

Then, it is highly desirable to reach an "ON" state for spin transmission in the same device. Since the inverse spin Hall effect in Pt can only detect the spin with an in-plane orientation, a nonzero component of the Néel vector in Cr_2O_3 in the sample plane is essential for measurable spin transmission for the present device. From the calculation of the Néel vector orientation in a uniaxial antiferromagnet under an external magnetic field, we found that the N´eel vector can be tilted slightly when the external field is neither parallel nor perpendicular to it[23]. Guided by this understanding, we measured the temperature dependence of the spin Seebeck effect in the Pt/Cr_2O_3/YIG device under different magnetic fields with a 20-degree tilting angle relative to the sample normal, the results of which are plotted in Fig. 3d. With an increase in the field magnitude from 0.5 T to 2.5 T, the enhancement of the spin Seebeck voltage is observed due to the rotation of the Néel vector. The change ratio due to the magnetic field, Ratio $(T_s)_{@H} = (V_{SSE@H} - V_{SSE@0.5T})/V_{SSE@0.5T}$, is plotted in Fig. 3e, which exceeds 500% for the temperature regime just below the Néel point and demonstrates an "ON" state for spin transmission. A systematic field angle dependence of the V_{SSE} results can be found in the original paper, which supports the Néel vector rotation scenario[20]. The Néel vector direction-dependent spin transport in AFMI has also been reported in a Pt/hematite/Pt lateral structure[24].

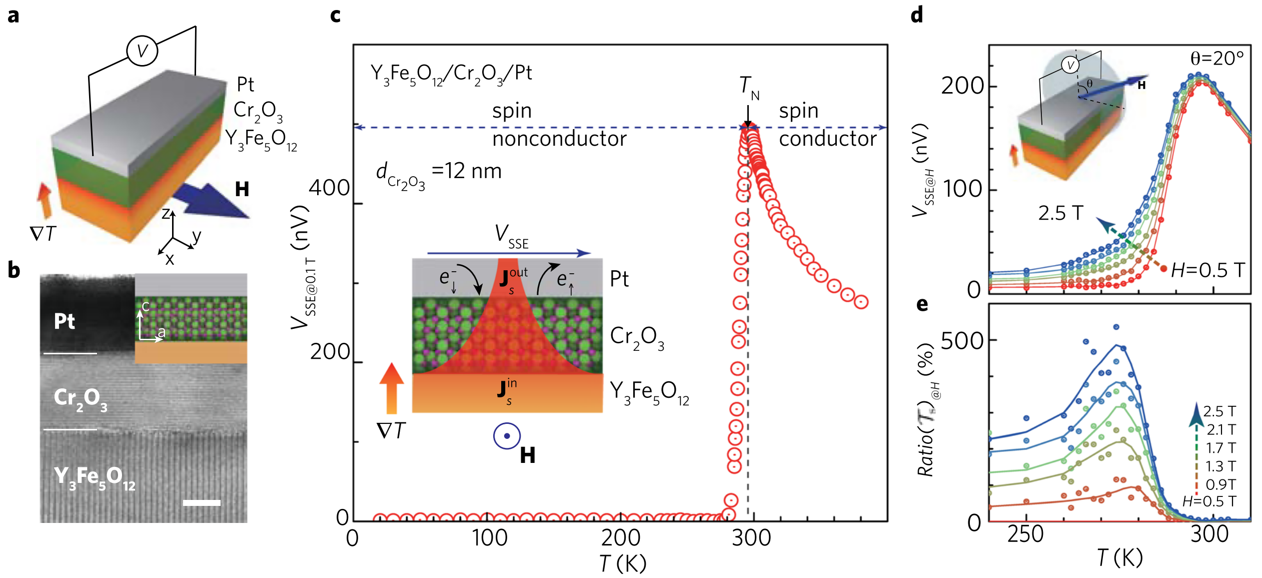

Fig. 3 **a** A schematic illustration of the spin Seebeck configuration for a YIG/Cr_2O_3/Pt trilayer device. A temperature gradient, ∇T, is along the *z* direction, while an external magnetic field (**H**) is along the *y* direction. **b** A cross-sectional TEM image of the YIG/Cr_2O_3/Pt trilayer device. The scale bar in the image is 5 nm. The out-of-plane direction *z* of the sample parallels the easy axis *c* of the Cr_2O_3. **c** The temperature dependence of the spin Seebeck voltage V_{SSE} at $H = 0.1$ T for the YIG/Cr_2O_3/Pt trilayer device. The inset shows the concept for measuring spin-current transmissivity. The magnetic field is aligned in the film plane. **d** Temperature dependence of V_{SSE} for the YIG/Cr_2O_3/Pt trilayer device at different external magnetic fields **H** at $\theta = 20°$. **e** The T_s change ratio Ratio $(T_s)_{@H}$ due to the external magnetic fields **H** as functions of temperature. Here, Ratio $(T_s)_{@H} = (V_{SSE@H} - V_{SSE@0.5T})/V_{SSE@0.5T}$. T_s refers to the spin-current transmissivity in the Cr_2O_3 layer

In addition to the recently studied spin transport, antiferromagnetic insulators also show a nontrivial effect on the magnetoresistance in neighboring heavy metals, such as Pt. In 2016, Shang et al. reported the temperature dependence of the magnetoresistance measurement in a Pt/NiO/YIG structure[25]. It was found that Pt shows a typical spin Hall magnetoresistance (SMR) behavior at room temperature, which was described for a Pt/YIG bilayer structure[26,27]. However, it is surprising that the SMR in Pt has a sign change for $T < 70$ K, which is hard to understand with the standard SMR model. Since the effect of NiO has been shown to quantitatively modulate the spin transmission between YIG and Pt, it is unexpected that the SMR shows a negative sign at low temperatures. We reproduced a sign change in a Pt/NiO/YIG device, and similar results were reported by another group[28]. The measurement configurations are illustrated in Fig. 4a–c, and the SMR results at 260 K and 20 K are plotted in Fig. 4d, e, respectively, which show the same SMR symmetry with opposite signs. Thus, the task is to explain the sign change. To achieve this, the magnetoresistance is measured for the Pt/NiO/YIG samples in a wide range of NiO thicknesses from 1.6 nm to 30 nm[29]. We noticed that the negative SMR is still finite even when the spin transmission between Pt and YIG is completely blocked for low temperature limits and thick NiO, indicating that the negative SMR is not caused by the spin current reflected from the YIG. Based on this observation, we develop the following interpretation, as illustrated in Fig. 3f. We attribute the negative SMR at low temperature to the NiO, which is assumed to have a 90° coupling with the YIG, which is a so-called spin-flop coupling[30]. In other words, the Pt/NiO interface contributes a negative SMR because the Néel vector of NiO is always perpendicular to the magnetic field. It is worth noting that NiO shows parallel coupling with a ferromagnet in some cases and winds up in a domain under manipulation[31,32]. With increasing temperature, the spin transmission between the Pt and YIG through the NiO, which contributes the conventional positive SMR, is enhanced. Therefore, a sign change of the SMR occurs when these two contributions compensate for each other. Recently, the spin-flop coupling between the NiO and YIG was confirmed by Luan et al. by polarized neutron reflectometry[33].

The negative SMR in the Pt/NiO bilayer was also reported by several other groups[34,35]. These works open the possibility of using AFM insulators as memory materials since orthogonal orientations of the Néel vector can be electrically determined. The prototype memory devices based on Pt/NiO bilayer structures were recently demonstrated independently by two groups, the results of which are shown in Fig. 4g, h[36,37].

Finally, we would like to discuss the challenges and unresolved problems in the study of spin transport in

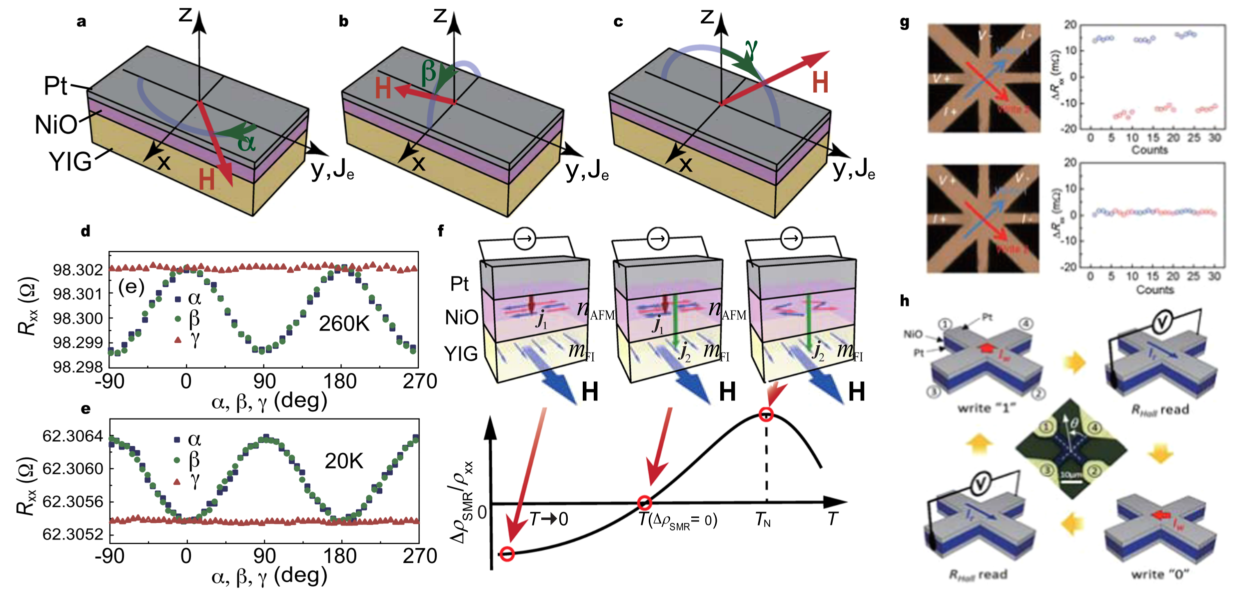

Fig. 4 a–**c** The illustrations for the magnetoresistance measurement configuration for magnetic field angle scan in different planes. Parameters *α*, *β*, and *γ* are the field angles defining the H directions when H is applied in the *x*–*y*, *x*–*z*, and *y*–*z* planes, respectively. **d**, **e** Field angle dependent resistance measured for the Pt/NiO/YIG device at 260 and 20 K with |H| = 20,000 Oe, which shows positive and negative SMR, respectively. **f** Illustrations for the interpretation of the sign change of the SMR in the Pt/NiO/YIG device; please see the main text for details. **g**, **h** SMR detection of the current-driven 90° rotation of the Néel vector in Pt/NiO/SrTiO$_3$(001) and Pt/NiO/Pt/MgO(111) structures. **g** Adapted from[36]. **h** Adapted from the ref. [37]

AFMIs. Although spin transport has been demonstrated in several AFMIs, the mechanisms of spin transfer through an AFMI, which is indispensable for further device design and development, have not been clarified. The scaling law of magnon spin current is another important issue to be addressed, which is closely related to the mechanism of spin transport. It might be approached by either a good theory capable of fitting the experimental data or a nicely designed experiment yielding the scaling law. Some unique features have been systematically captured by experiments but are still hard to understand, e.g., the pronounced microwave frequency dependence of spin pumping efficiency near the Néel point[15]. Meanwhile, the proposed spin transport mechanisms need to be carefully verified by some intelligent and well-controlled experiments[16,38]. Another challenge lies in excitation of the THz dynamics of an AFMI by a spin current, which is quite interesting but still challenging due to the large anisotropy of antiferromagnets[39–41].

Acknowledgements

This work was supported by JST-ERATO "Spin Quantum Rectification", JST-PRESTO "Phase Interfaces for Highly Efficient Energy Utilization", Grant-in-Aid for Scientific Research on Innovative Area, "Nano Spin Conversion Science" (26103005 and 26103006), Grant- in-Aid for Scientific Research (S) (25220910), Grant-in-Aid for Scientific Research (A) (25247056 and 15H02012), Grant-in-Aid for Challenging Exploratory Research (26600067), Grant-in-Aid for Research Activity Start-up (25889003), and World Premier International Research Center Initiative (WPI), all from MEXT, Japan. Z.Q. acknowledges support from the "Fundamental Research Funds for the Central Universities (DUT17RC(3)073)".

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Conflict of interest

The authors declare that they have no conflict of interest.

Publisher's note

Received: 21 September 2018 Revised: 15 February 2019 Accepted: 14 April 2019.

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