

OPEN

Anomalous anisotropic behaviour of spin-triplet proximity effect in Au/SrRuO₃/Sr₂RuO₄ junctions

M. S. Anwar¹ ^{2*}, M. Kunieda¹, R. Ishiguro^{3,4}, S. R. Lee^{5,6}, C. Sow¹, J. W. A. Robinson², S. Yonezawa¹, T. W. Noh¹ ^{5,6} & Y. Maeno¹

Spin-polarized supercurrents can be generated with magnetic inhomogeneity at a ferromagnet/spin-singlet-superconductor interface. In such systems, complex magnetic inhomogeneity makes it difficult to functionalise the spin-polarized supercurrents. However, spin-polarized supercurrents in ferromagnet/spin-triplet-superconductor junctions can be controlled by the angle between magnetization and spin of Copper pairs (*d*-vector), that can effectively be utilized in developing of a field of research known as superconducting spintronics. Recently, we found induction of spin-triplet correlation into a ferromagnet SrRuO₃ epitaxially deposited on a spin-triplet superconductor Sr₂RuO₄, without any electronic spin-flip scattering. Here, we present systematic magnetic field dependence of the proximity effect in Au/SrRuO₃/Sr₂RuO₄ junctions. It is found that induced triplet correlations exhibit strongly anisotropic field response. Such behaviour is attributed to the rotation of the *d*-vector of Sr₂RuO₄. This anisotropic behaviour is in contrast with the vortex dynamic. Our results will stimulate study of interaction between ferromagnetism and unconventional superconductivity.

Generation of dissipationless spin-polarized (spin-triplet) supercurrent is the major interest of superconducting devices, which can be utilized to establish energy efficient superconducting spintronics^{1,2}. In last two decades, rigorous works have been conducted to produce and control spin-triplet supercurrents using heterostructures of ferromagnets (Fs) and conventional spin-singlet superconductors (SSCs)^{3–13}. It has been established that magnetic inhomogeneity is always required to emerge spin-triplet supercurrent at F/SSC interface, which can be achieved by e.g. using non-collinear magnetization in multilayer ferromagnets^{6,7,9}. Complicated magnetic structure of multilayer ferromagnets makes it hard to functionalize the F/SSC devices. This issue can be settled by replacing SSC with a spin-triplet superconductor (TSC). In F/TSC heterostructures, a single F layer can effectively emerge spin-polarized supercurrents with fully conserved spin degree of freedom in the entire device. Furthermore, recent theoretical work suggested that superconducting properties of F/TSC junctions strongly depends on relative orientation of magnetization (*m*) of the F and *d*-vector of the TSC^{14–18}. Interestingly, when the *d*-vector and *m* are perpendicular (i.e. when the angle between *d*-vector and *m*, $\theta_{md} = \pi/2$) the spin-triplet correlation can induce monotonically over a long range. However, for parallel configuration, i.e. $\theta_{md} = 0$, the induced order parameter oscillates spatially¹⁸.

Sr₂RuO₄ (SRO214) is one of the best-candidates of TSCs¹⁹ with the superconducting critical temperature (*T*_c) of 1.5 K. Most likely, it exhibits the chiral *p*-wave spin-triplet state with broken time-reversal symmetry^{20–28}, although there are still unresolved issues^{29–31}. We should comment that the very recent nuclear magnetic resonance (NMR) study shows the reduction of Knight shift, which cannot be explained by a simple chiral-*p*-wave spin-triplet scenario^{32,33}. Such a chiral-*p*-wave superconductivity in SRO214 attracts interest for exploring topological superconducting phenomena originating from its orbital phase winding^{19,26–28}. The superconducting order parameter of a SRO214 can be represented by a vector $d = \hat{z}(p_x \pm ip_y)$, where \hat{z} is the out-of-plane (along the *c*-axis) basis vector, and *p*_{*x*} and *p*_{*y*} are the *x* and *y* components of the orbital order parameters, respectively¹⁹. Note that the *d*-vector is always perpendicular to the spin of triplet Cooper pairs. For bulk SRO214 superconductor,

¹Department of Physics, Kyoto University, Kyoto, 606-8502, Japan. ²Department of Materials Science and Metallurgy, University of Cambridge, Cambridge, UK. ³Department of Mathematical and Physical Sciences, Japan Women's University, Tokyo, 112-8681, Japan. ⁴Department of Applied Physics, Tokyo University of Science, RIKEN, Tokyo, 162-8601, Japan. ⁵Center for Correlated Electron Systems, Institute for Basic Science (IBS), Seoul, 151-747, Republic of Korea. ⁶Department of Physics and Astronomy, Seoul National University, Seoul, 151-747, Republic of Korea. *email: msa60@cam.ac.uk

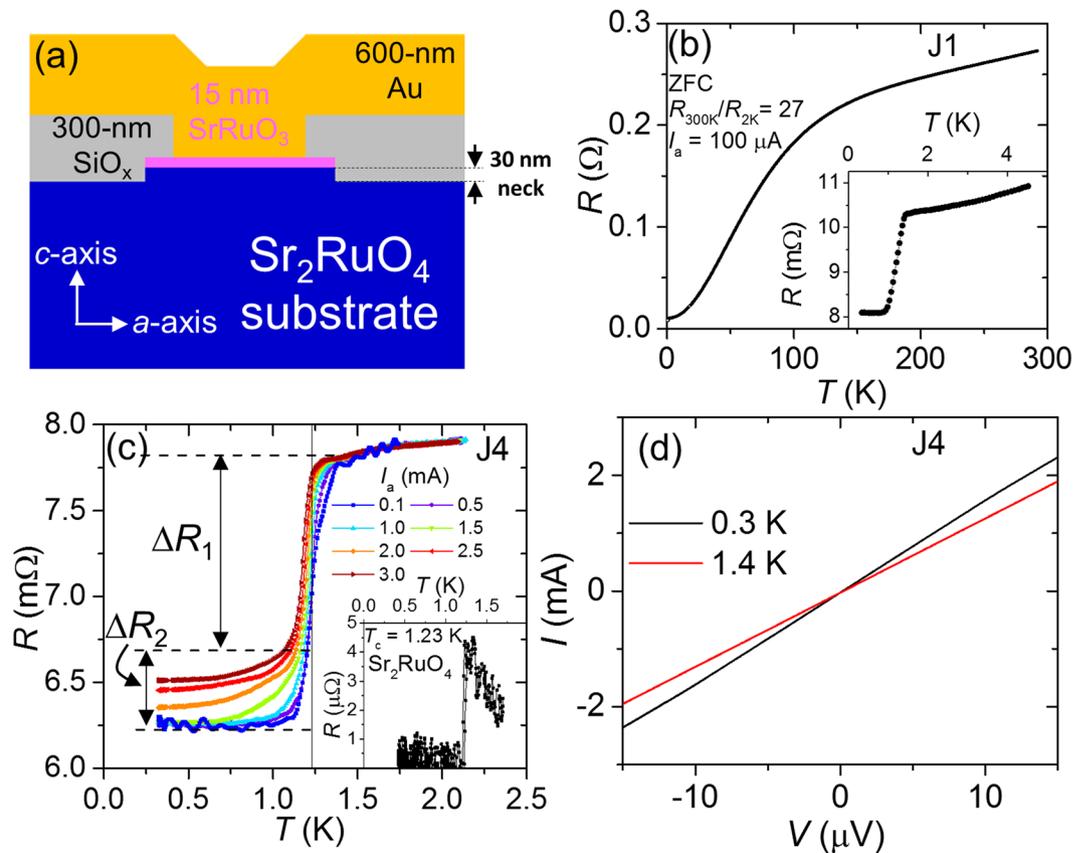


Figure 1. (a) Schematic of Au/SrRuO₃/Sr₂RuO₄ junctions. Note that a ≈ 30 -nm thick neck of SRO214 was prepared in order to separate a part of superconductor from the bulk substrate. (b) Temperature dependent resistance $R(T)$ from 300 K down to 2 K for Junction J1 with the junction area of $20 \times 20 \mu\text{m}^2$. Inset shows $R(T)$ close to the superconducting transition. (c) $R(T)$ of Junction J4 ($5 \times 5 \mu\text{m}^2$) at various applied currents. Vertical solid line indicates the bulk T_c of SRO214. Inset shows $R(T)$ of the SRO214 substrate measured by using a four probes technique. (d) Current-voltage curves below and above T_c of Junction J4.

d -vector is aligned along the c -axis (out-of-plane) fixed by spin-orbit coupling^{34–36} but at the surface it may not be true because of surface effects. It is expected that d -vector may rotate to the in-plane direction with external magnetic field of about 20 mT applied along the out-of-plane direction^{37,38}.

Newly, we developed F/TSC heterostructures by growing epitaxial ferromagnetic SrRuO₃ (SRO113) thin films on a spin-triplet superconductor SRO214 single crystal using pulsed laser deposition³⁹. Furthermore, long-ranged proximity effect is also observed, where even frequency p -wave spin-triplet correlation may dominate compared to odd frequency s -wave spin-triplet, as junctions were in clean limit^{40,41}.

In this article, we present our investigations on differential conductance (dI/dV) of Au/SRO113/SRO214 junctions as a function of temperature and magnetic field applied along both in-plane (H_{in}) and out-of-plane (H_{out}) directions. We found that the induced correlation exhibits anisotropic behaviour in the response of applied field. Vortex dynamics cannot explain our results. This anisotropic effect can be attributed to the relative orientation of the d -vector of SRO214 and m of SRO113.

Results

In this article, we present the results of various junctions but mainly focus on two junctions J4 and J5 that exhibit different normal-state resistance (R_N) 7.83 m Ω and 198 m Ω , respectively. A schematic illustration of a junction is shown in Fig. 1(a).

Temperature dependent resistance $R(T)$ was measured during cooling in zero field down to 300 mK. SRO214, SRO113 and Au are good metals with very low resistivity at low temperatures, except the resistivity of SRO214 along the c -axis (ρ_c)²⁹. It suggests that at low temperatures resistivity of our junctions is dominated by the SRO113/SRO214 and Au/SRO113 interfaces^{39,40}. Our present junctions exhibit RRR = 27, which that is three times larger than that of our previous junctions⁴⁰. Furthermore, a sharp superconducting transition is observed that reflects good quality of the junctions (inset of Fig. 1(b)). It is also clear that, in the normal state, the resistivity data have major contributions of ρ_c of SRO214, as evidenced by the characteristic hump in $R(T)$ at around 100 K. This result indicates that current is flowing along the normal of the junctions, which excludes the possibility of any direct contact between Au electrode and SRO214 superconductor.

Figure 1(c) presents $R(T)$ at low temperatures of Junction J4 ($5 \times 5 \mu\text{m}^2$) measured with different applied currents (I_a in the range of 0.1 to 3 mA) that shows two superconducting regions robust and weak against I_a . At

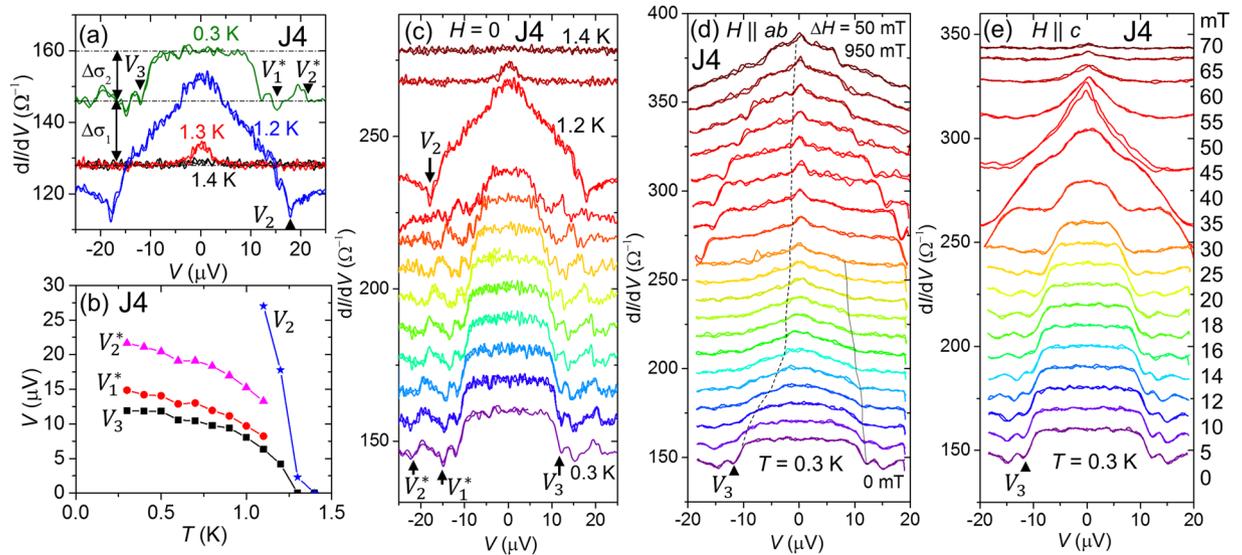


Figure 2. Temperature and field dependent differential conductance (dI/dV) of Au/SrRuO₃/Sr₂RuO₄ Junction J4. **(a)** dI/dV measured at different temperatures between 1.4 K and 0.3 K. At 0.3 K a flat-top central conductance peak appears between $\pm V_3$. Note that there are additional oscillations with the minima at V_1^* and V_2^* . At temperatures above 1.1 K, another transition at $\pm V_2$ becomes visible within the range of bias voltage. **(b)** Temperature dependence of all characteristic voltages appeared in dI/dV data. **(c)** dI/dV at various temperatures. The data are shifted with a step of $10 \text{ m}\Omega^{-1}$ but for 1.2 K and 1.3 K are shifted with $30 \text{ m}\Omega^{-1}$ for clarity. dI/dV as a function of magnetic field applied along **(d)** the in-plane and **(e)** the out-of-plane (c -axis) directions. Data are shifted for clarity.

$I_a = 0.1 \text{ mA}$ the transition onset appears at $T_c = 1.38 \text{ K}$, which is higher than $T_c \approx 1.23 \text{ K}$ of SRO214 substrate (see the inset of Fig. 1(c)). This increase in T_c may arise due to mutual strain (pressure) between the substrate and the film close to the interface⁴². The $R(T)$ behaviour with increasing I_a reveals that there are two distinct transition regions. The first transition with change in the resistance $\Delta R_1 = 1.15 \text{ m}\Omega$ is only weakly dependent on I_a . The second transition is strongly suppressed with increasing I_a with the change in the resistance of $\Delta R_2 = 0.45 \text{ m}\Omega$. Multiple transitions are expected to emerge in a multi-barrier junction such as the junctions presented here: Normal-metal/F/TSC. Essentially, the same behaviour has been observed in our previous study⁴⁰. These observations suggest that ΔR_1 and ΔR_2 correspond to resistance changes at the SRO113/SRO214 and Au/SRO113 interfaces, respectively. It means that dI/dV as a function of bias voltage should exhibit Andreev reflection (AR) features occurring at two distinct interfaces.

Note that AR can occur for a bias voltage below superconducting gap (induced minigap in SRO113⁴¹) at SRO113/SRO214 (Au/SRO113) interface. To study the AR, we obtained $dI/dV(V)$ by taking first order numerical derivative of the measured current-voltage (I - V) curves shown in Fig. 1(d). We investigate dI/dV as a function of externally applied magnetic fields both in-plane H_{in} and H_{out} directions and temperature as well.

First, we present dI/dV data measured at different temperatures (see Fig. 2(a)). It shows an enhancement in dI/dV below superconducting transition temperature T_c . Below T_c , the conductance of a transparent F/SSC interface increases with opening of AR channel, which is related with spin-polarization of the ferromagnet as well. At 0.3 K and zero applied field (after zero field cooling), the conductance enhances by $\approx 10\%$ below $\pm V_3$ with almost flat top. This transition yields the change in the conductance $\Delta\sigma_2 = 13.84 \text{ }\Omega^{-1}$ that corresponds to ΔR_2 (second transition in $R(T)$). At temperatures higher than 1.1 K an additional enhancement within our applied bias voltage can be observed at $\pm V_2$. Note that there are sharp dips associated with V_2 features (Fig. 2(a)), which may correspond to the destruction of superconductivity at SRO113/SRO214 interface with critical current⁴¹. However, the temperature dependence of dI/dV (Fig. 2(c)) shows that the dip persists up to the bulk T_c , which suggests that the dip can be attributed to the critical current of SRO214-neck part of the junctions, and the proximity effect at the SRO113/SRO214 interface is simultaneously switched off. The V_2 transition exhibits $\approx 14\%$ increase with $\Delta\sigma_1 = 18.04 \text{ }\Omega^{-1}$ consistent with ΔR_1 (first transition in $R(T)$). It indicates that characteristic voltages of V_3 and V_2 are corresponding to multi AR features occurred at Au/SRO113 and SRO113/SRO214 interfaces, respectively. The data presented in Fig. 2(b) is extracted from Fig. 2(c). It shows a monotonic suppression of the AR with increase in the temperature. Similar behaviour is observed for junction J5 as well which is shown in Fig. 3(f) and discussed more at the end of the section.

The observations of multiple enhancements in dI/dV reveal that superconductivity penetrates into the 15-nm hick SRO113 layer and reaches at the Au/SRO113 interface as well. This is only possible with spin-triplet superconducting correlation, since spin-singlet coherence length of SRO113 is of the order of 1-nm⁴³. Note that two additional oscillations with minima at V_1^* and V_2^* appear between V_3 and V_2 . These oscillations are suppressed monotonically just like V_3 with the increase in the temperature, see Fig. 2(b,c). These oscillations may correspond to McMillan-Rowell resonance⁴⁴ resulting from interference between Andreev reflected quasiparticles at the SRO113/SRO214 interface and the quasiparticles reflected back into SRO113-layer from the Au/SRO113 interface.

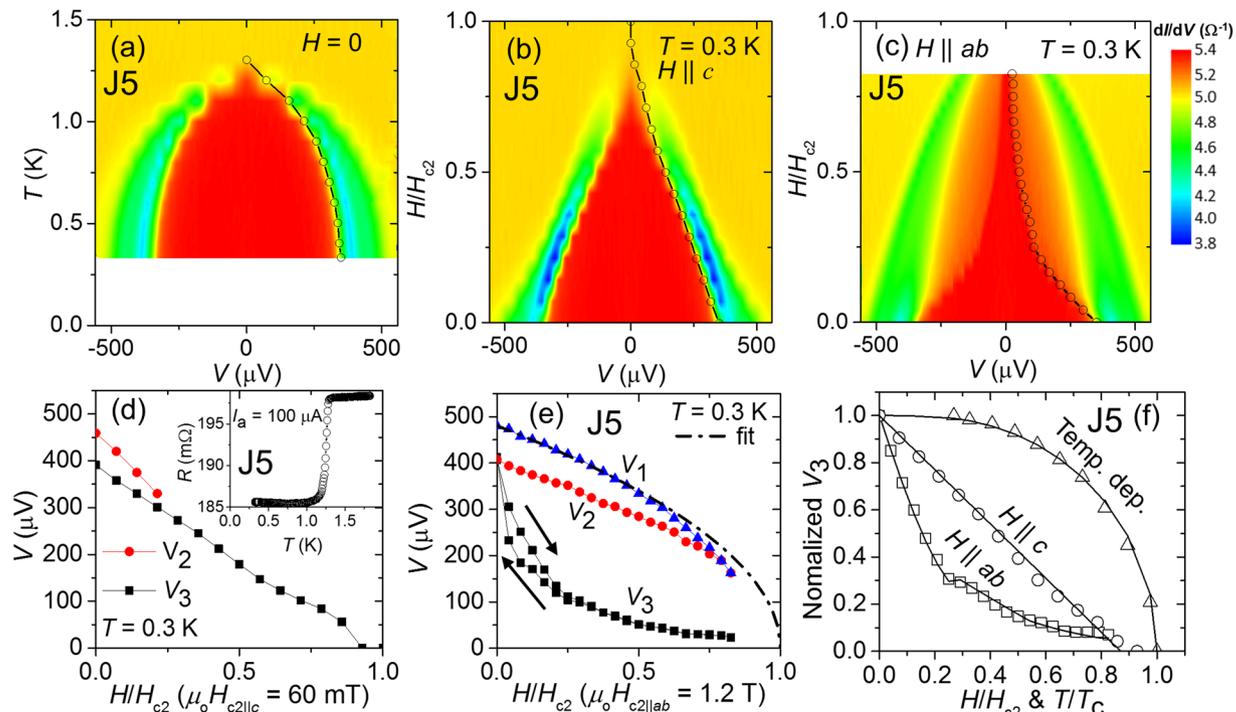


Figure 3. Differential conductance of Au/SrRuO₃/Sr₂RuO₄ Junction J5. (a) Colour plot of $dI/dV(V)$ vs temperature. The V_3 is indicated with open circles. $dI/dV(V)$ vs magnetic field applied along (b) the c -axis and (c) the in-plane directions, respectively. (d) V_2 and V_3 vs applied field along the c -axis normalized by the upper critical field. Inset shows $R(T)$ at low temperatures of Junction J5. (e) All characteristic voltages V_1 , V_2 and V_3 vs in-plane applied field. V_1 and V_2 decrease expectedly. In contrast V_3 exhibits an anisotropic behaviour with hysteresis below 200 mT. (f) Normalized V_3 vs applied field along both in-plane (H_{in}) and out-of-plane directions H_{out} , as well as vs temperature plotted for comparison.

The most interesting and important part of this study is voltage biased dI/dV as a function of applied field. Figure 2(d,e) shows the $dI/dV(V)$ data obtained with the fields H_{in} and H_{out} , respectively. In case of the applied field H_{in} , V_3 is abruptly suppressed up to 200 mT and decreases slowly with further increase in H_{in} . In contrast V_2 is suppressed gradually, which is an expected behaviour of Cooper pair breaking under applied magnetic fields (orbital effects). For fields H_{out} , both V_3 and V_2 decreases monotonically. Thus spin-triplet proximity effect into the SRO113 layer exhibits an anisotropic response to applied field. Such an effect can be expected for anisotropic d -vector rotation with applied field, further discussions are given in the discussions section.

Junction J4 with lower R_N exhibits various interesting features in the junction voltage. However, concerning to the signal to noise ratio a junction with larger R_N is preferable. Indeed in Junction J5 with rather high $R_N = 198.2$ m Ω , we observed good quality data that also exhibits AR features with multiple transitions. For Junction J5, $dI/dV(V)$ as a function of temperature and applied field exhibit two transitions corresponding to V_3 and V_2 . Note that we also obtained the V_1 feature particularly at 0.3 K and zero field as observed in our previous junctions⁴⁰. The characteristic voltage V_1 most probably arises due to critical-current of SRO214 neck⁴⁰. Note; in this junction the oscillations corresponding to McMillan-Rowell resonance are not observed due to its lower amplitude and higher R_N . By comparing Fig. 3(b,c), a strong anisotropy in V_3 is obviously reproduced. In fact, we observed this anisotropic effect in almost all our working junctions.

For Junction J5, under the effect of H_{out} fields, the characteristic voltage V_2 shown in blue in Fig. 3(b) becomes vague at higher fields as shown in Fig. 3(d). It reflects that in junctions with higher R_N (lower interface conductance) the AR features are overlapped with each other. However, all the characteristic-voltage features are obviously present for all H_{in} fields, see Fig. 3(e). Interestingly, V_3 exhibits hysteretic effect only below 200 mT. For a comparison, we plotted normalized V_3 as a function of field and temperature in Fig. 3(f).

We applied theoretical fits on temperature and field dependent V_3 to analyse its behaviour. The behaviour of V_3 versus temperature follows the interpolation formula, $V_3(T) = V_3(0) \tanh \sqrt{a(T_c/T - 1)}$ with constant $a = 1.56$. We comment that the formula with the same functional form can be used to approximate the temperature dependence of the superconducting gap, with $a = 1.74$ for the s -wave superconductivity. Interesting, $a = 1.56$ indicates unconventional superconducting proximity effect and potentially consistent with p -wave superconductivity^{41,45}. The monotonic suppression of V_3 under fields H_{out} can be reasonably fitted with the expression $V_3(H) = V_3(0) \sqrt{1 - H/H_c}$. However, V_3 under fields H_{in} is strongly suppressed at lower fields less than 200 mT and slowly decreases for higher fields. Note that the coercive field of SRO113 layer is ≈ 200 mT for rectangular pads of the size of tens of microns⁴⁰. It indicates that there are two distinct decoherence effects on the induced spin-triplet correlation in the SRO113 layer, such as orbital effect and m rotation relative to the d -vector of SRO214 superconductor. The phenomenological fit shown in Fig. 3(f) is discussed below.

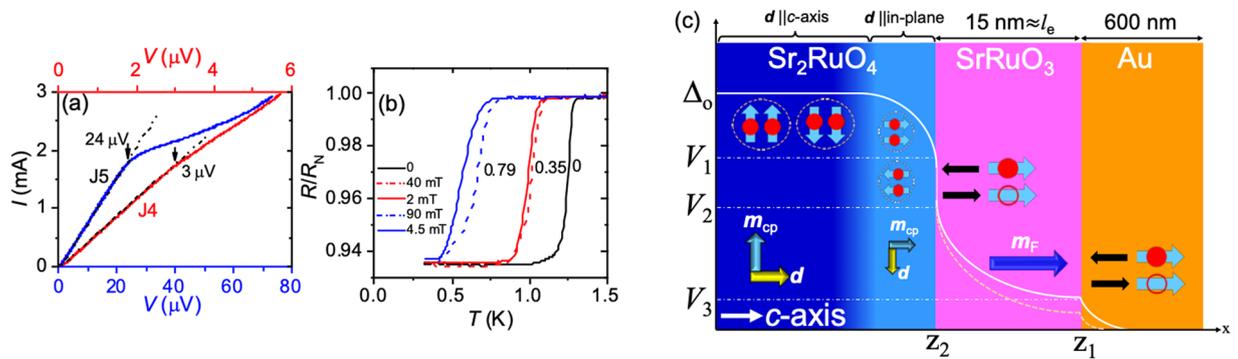


Figure 4. (a) Current-voltage curves at 0.3 K for Junctions J4 (red) and J5 (blue) after subtracting additional contributions. (b) $R(T)$ at different applied fields both H_{in} and H_{out} . (c) Schematic illustration of induction of superconductivity in the junction. Explanations are given in the main text.

Discussion

Before starting our discussion, let us summarize our main results. We observed AR features in dI/dV with characteristic voltages V_3 and V_2 emerging at Au/SRO113 and SRO113/SRO214 interfaces, respectively. The V_3 (V_2) exhibits anisotropic (isotropic) behaviour in the response of applied magnetic field. The AR feature with V_3 is suppressed strongly below 200 mT and decreases slowly with further increase under the field H_{in} . However, V_2 decreases monotonically with H_{out} and H_{in} as well. For the field along the c -axis V_3 decreases similar to V_2 .

First, we discuss the subtraction of resistance contributions other than interfaces to estimate the accurate values of characteristic voltages V_3 and V_2 . As described below, the measured resistance of our devices may contain some contributions from non-interface resistance, such as from the neck part of SRO214. To estimate accurate values of characteristic voltages specifically, V_3 at 0.3 K, we subtract the contributions of estimated additional resistances. For simplicity, let us assume an ideal situation that AR probability is 100% so that the conductance of each interface is enhanced by a factor of two compared to its normal-state conductance. Under this assumption, for Junction-J4 (JunctionJ5) the total resistance of Au/SRO113 and SRO113/SRO214 interface is two times the $\Delta R = \Delta R_1 + \Delta R_2 = 1.55$ m Ω (12.74 m Ω). At 0.3 K, additional resistance of JunctionJ4 (JunctionJ5) $R_a = R_N - 2\Delta R = 4.73$ m Ω (172.72 m Ω) that contributes additional voltage drop across the junction. We subtract a linear contribution of R_a and plotted the results in Fig. 4(a). It reveals that V_3 for JunctionJ4 (junction-J5) is ≈ 3 μ V (≈ 20 μ V). It shows that JunctionJ5 exhibits an order higher V_3 due to lower interface transparency and more drop of voltage across it.

It is important to discuss the vortex effect on proximity effect in Au/SRO113/SRO214 junctions, which can also be anisotropic. Of course, vortices can be induced for the fields higher than H_{c1} . Note that H_{c1} of SRO214 is 1 mT and 70 mT along the a -axis (in-plane) and the c -axis¹⁹, respectively. Transport properties of a superconducting junction can be affected when vortices are crossing the interface because of vortex dynamics. According to the geometry of our junctions (c -axis is normal of the junction), the field induced vortices can encounter the interface only for fields H_{out} . However, vortices align parallel to the interface for the fields H_{in} . It means, AR features should exhibit additional suppression for fields H_{out} . In contrast, V_3 is suppressed strongly for the fields H_{in} only.

To understand the effect of vortices in our junctions, we measured $R(T)$ under the fields H_{in} and H_{out} . Even for the same effective field, T_c of the first transition with ΔR_1 of SRO113/SRO214 interface decreases more under H_c than H_{ab} , see Fig. 4(b). However, the second transition corresponding to Au/SRO113 interface is in contrast. It indicates that the electronic transport properties of our junctions can encounter vortices only for the fields H_{out} . Furthermore, it also reveals that the observed anisotropic response of V_3 under applied fields does not arise due to vortex dynamics.

Effect from the spin part: spin-triplet proximity effect at F/TSC interface can be dictated by angle θ_{dm} between d -vector of SRO214 and m vector of SRO113 layer^{14–18}. We applied a following phenomenological fit to $V_3(H_{in})$,

$$V_3(H) = V_3(0)[F(H) - G(\theta_{md})] \quad (1)$$

where, $V_3(0)$ is the characteristic voltage at zero temperature, $F(H) = \sqrt{1 - H/H_{c2}}$ and $G(\theta_{md}) = A \cos(\theta_{md})$ with a constant A . We obtained a good fit by changing the θ_{md} nonmonotonically; first linearly decreased until 200 mT and increased for higher fields. This model implicitly assumes that that d -vector is aligned along the in-plane direction at the interface.

In bulk SRO214 superconductor, the d -vector is aligned along the c -axis and in the result the spins of the Cooper pairs are aligned along the ab -plane. The d -vector and Cooper pair spins can be rotated with an applied magnetic field H_{out} of 20 mT^{37,38} to in-plane and to out-of-plane, respectively. In this scenario, H_{in} cannot rotate the d -vector. That means, proximity effect should suppress more in case of H_{out} rather than H_{in} . But, our results are suggesting in contrast.

If the d -vector is perpendicular a space between perpendicular and to the c -axis particularly at the interface, V_3 can be affected with rotation of θ_{md} with H_{in} . It is possible that broken inversion symmetry at the SRO113/SRO214 interface induces Rashba-type⁴⁶ spin-orbit coupling with in-plane vector characterizing the spin-orbit coupling $g = (k_y, k_x, 0)$ with out-of-plane effective field, which tends to align the d -vector along the in-plane direction. Additionally, out-of-plane magnetic anisotropy of SRO113 thin film is also supporting such an alignment of

the d -vector. As a result, at zero applied field, spin of triplet Cooper pairs in SRO214 and m of SRO113 are parallel and thus $\theta_{md} = \pi/2$, which is a favourable configuration for proximity effect in F/TSC junctions. The angle θ_{md} can be reduced with either rotation of m or d -vector that dictates the proximity effect in the junction accordingly¹⁸. It is most likely that the rotation of m of SRO113 is responsible for first strong suppression in V_3 up to coercive field of SRO113-layer ≈ 200 mT⁴⁰. It may also be the reason of hysteresis in V_3 below 200 mT; above 200 mT, m is saturated and fully aligned along H_{in} . As a result θ_{md} decreases up to coercive field and suppresses the proximity effect. On the other hand, at the same time, the applied field H_{in} tends to align the d -vector along the c -axis. Therefore, θ_{md} never becomes zero and proximity effect is weakly suppressed with increasing H_{in} above 200 mT. In this configuration of d -vector at the interface as illustrated in Fig. 4(c), H_{out} cannot rotate the d -vector, as spins of Cooper pairs are aligned to the out-of-plane direction. Therefore, V_3 suppresses monotonically with H_{out} . Of course, temperature change between 2 K and 0.3 K cannot change m or d -vector. Thus, there is no anomalous suppression in V_3 as a function of temperature.

The rotation of the d -vector under the influence of an external magnetic field is well known for superfluid³ He-A phase⁴⁷ analogous to the chiral p -wave spin-triplet superconductivity anticipated in SRO214. In case of the standard chiral- p -wave scenario for SRO214, the d -vector should be fixed along the c -axis because of spin-orbit coupling. Possibility of rotation of the d -vector to an in-plane direction under c -axis magnetic field is discussed in the NMR study for $H||c$ ³⁷. However, at SRO113/SRO214 interface, broken inversion symmetry and out-of-plane exchange field of SRO113 can significantly affect the spin-orbit coupling. That may result into easier rotation of the d -vector along the in-plane direction. This scenario explains our results well. Theoretically, Annett *et al.*³⁸, predicted that the d -vector rotation by magnetic field along the c -axis is accompanied by chiral to nonchiral phase transition depending on spin dependent effective pairing interaction. However, for a proximity effect in a junction based on a spin-triplet superconductor, the crucial parameter is the orientation of the d -vector¹⁸, whereas, the transition in the orbital part may not considerably modulate the proximity effect unless the junction size is of the order of the typical size of a chiral domain.

Effect from the orbital part: SRO214 exhibits multi-component ($p_x + ip_y$) superconducting order parameter²⁸. That may also explain this anisotropic behaviour of V_3 . The component p_x can be significantly suppressed compared to p_y when magnetic field H_{in} is parallel to y and vice versa^{48–50}. However, the fields along the z -direction, perpendicular to the both p_x and p_y components, can simultaneously affect both components of the order parameter. In this way, the differential conductance should depend differently on the in-plane and out-of-plane applied magnetic fields. Such anisotropic effect corresponding to the order parameter of the bulk SRO214 should emerge mainly in the SRO214 side and that should result in anisotropic behaviour either only in V_2 or both V_2 and V_3 . Thus, our observed anomalous anisotropic behaviour only in V_3 cannot be explained by the above mentioned non-chiral scenario. Rather, this observation supports that the spin-triplet proximity effect is controlled with the change in θ_{md} due to the relative rotation of m and d -vector. More experimental and theoretical works are needed to understand and manipulate the rotation of θ_{md} .

Conclusion

We studied long-range proximity effect in Au/SRO113/SRO214 double barrier junctions. Two distinct Andreev reflection features are observed in differential conduction vs bias voltage coming from two SRO113/SRO214 and Au/SRO113 interfaces with the induction of spin-triplet Cooper pairs in 15-nm thick SRO113 from SRO214. The transition of Au/SRO113 interface is suppressed anisotropically with externally applied magnetic fields. This anomalous anisotropic behaviour cannot be explained with vortex dynamics since differential conductance as a function of temperature and out-of-plane applied fields is qualitatively similar. However, conductance of SRO113/SRO214 interface suppressed faster when field is applied along the in-plane direction. Our results will stimulate the theoretical work to understand the correlation between unconventional superconductivity and ferromagnetism, p -wave proximity effect and applications to initiate the Superconducting Spintronics.

Methods

Single crystals of SRO214 were grown in Kyoto using a floating-zone method⁵¹. Some parts of the SRO214 crystals tend to contain $\text{Sr}_3\text{Ru}_2\text{O}_7$, SRO113, as well as Ru-metal inclusions. These inclusions are unavoidable in order to eliminate Ru deficiencies and obtain T_c close to the intrinsic $T \approx 1.5$ K. To fabricate our superconducting junctions, we carefully choose the parts of SRO214 crystals that do not contain impurities but with a slightly lower $T_c \approx 1.23$ K. Ferromagnetic SRO113 thin films with thickness of 15-nm were epitaxially deposited using pulsed laser deposition on the cleaved ab -surface of the SRO214 substrates ($3 \times 3 \times 0.5$ mm³) in Seoul. The details of SRO113 thin film deposition can be found elsewhere^{39,40}. Immediately after the growth of SRO113 film, a 5-nm thick Ti adhesive layer and a 20-nm thick Au capping layer were deposited *ex-situ* by dc-sputtering.

Double interface Au/SRO113/SRO214 junctions shown schematically in Fig. 1a were fabricated in RIKEN. First, 25×25 μm^2 and 10×10 μm^2 pads of SRO113 were prepared on flat surfaces using photolithography and Ar ion etching. Then, an insulating 300-nm thick SiO_x layer was sputtered. Finally, a 600-nm thick Au top electrode was deposited by electron beam evaporation with junction areas of 20×20 μm^2 and 5×5 μm^2 over 25×25 μm^2 and 10×10 μm^2 SRO113 pads, respectively. Note, a 30-nm thick neck part of SRO214 substrate was prepared with over-etching during SRO113 layer etching to define a bottom superconducting electrode, as shown in Fig. 1(a). Detailed descriptions of these processes are given in ref.⁴⁰.

Electrical transport measurements were performed using the four-point technique with two contacts (I_+ , V_+) on the Au top electrode and the other two contacts (I_- , V_-) connected directly to the side of the SRO214 crystal. Transport properties were studied down to 300 mK using a ³He cryostat equipped with a vector superconducting magnet.

Received: 5 June 2019; Accepted: 10 October 2019;

Published online: 01 November 2019

References

- Eschrig, M. Spin-polarized supercurrents for spintronics: a review of current progress. *Rep. Prog. Phys.* **78**, 104501 (2015).
- Linder, J. & Robinson, J. W. A. Superconducting spintronics. *Nat. Phys.* **11**, 307 (2015).
- Bergeret, F. S., Volkov, A. F. & Efetov, K. B. Long-range proximity effects in superconductor-ferromagnet structures. *Phys. Rev. Lett.* **86**, 4096 (2001).
- Eschrig, M. & Löfwander, T. Triplet supercurrents in clean and disordered half-metallic ferromagnets. *Nat. Phys.* **4**, 138 (2008).
- Keizer, R. S. *et al.* A spin triplet supercurrent through the half-metallic ferromagnet CrO₂. *Nature(London)* **439**, 825 (2006).
- Khaire, T. S., Khasawneh, M. A., Pratt, W. P. Jr. & Birge, N. O. Observation of spin-triplet superconductivity in Co-based Josephson junctions. *Phys. Rev. Lett.* **104**, 137002 (2010).
- Robinson, J. W. A., Witt, J. D. S. & Blamire, M. G. Controlled injection of spin-triplet supercurrents into a strong ferromagnet. *Science* **329**, 59 (2010).
- Anwar, M. S., Czeschka, F., Hesselberth, M., Porcu, M. & Aarts, J. Long-range supercurrents through half-metallic ferromagnetic CrO₂. *Phys. Rev. B* **82**, 100501(R) (2010).
- Anwar, M. S., Veldhorst, M., Brinkman, A. & Aarts, J. Long range supercurrents in ferromagnetic CrO₂ using a multilayer contact structure. *Appl. Phys. Lett.* **100**, 052602 (2012).
- Bergeret, F. S. & Tokatly, I. V. Singlet-Triplet Conversion and the Long-Range Proximity Effect in Superconductor-Ferromagnet Structures with Generic Spin Dependent Fields. *Phys. Rev. Lett.* **110**, 117003 (2013).
- Bergeret, F. S. & Tokatly, I. V. Spin-orbit coupling as a source of long-range triplet proximity effect in superconductor/ferromagnet hybrid structures. *Phys. Rev. B* **89**, 134517 (2014).
- Jacobsen, S. H., Kulagina, I. & Linder, J. Controlling superconducting spin flow with spin-flip immunity using a single homogeneous ferromagnet. *Sci. Rep.* **6**, 23926 (2016).
- Banerjee, N. *et al.* Controlling the superconducting transition by spin-orbit coupling. *Phys. Rev. B* **97**, 184521 (2018).
- Brydon, P. M. R. & Manske, D. 0- π Transition in Magnetic Triplet Superconductor Josephson Junctions. *Phys. Rev. Lett.* **103**, 147001 (2009).
- Brydon, P. M. R. Spontaneous spin current due to triplet superconductor-ferromagnet interfaces. *Phys. Rev. B* **80**, 224520 (2009).
- Annunziata, G., Cuoco, M., Noce, C., Sudbø, A. & Linder, J. Spin-sensitive long-range proximity effect in ferromagnet/spin-triplet-superconductor bilayers. *Phys. Rev. B* **83**, 060508(R) (2011).
- Gentile, P. *et al.* Spin-orbital coupling in a triplet superconductor-ferromagnet junction. *Phys. Rev. Lett.* **111**, 097003 (2013).
- Terrade, D., Gentile, P., Cuoco, M. & Manske, D. Proximity effects in a spin-triplet superconductor-ferromagnet heterostructure with a spin-active interface. *Phys. Rev. B* **88**, 054516 (2013).
- Maeno, Y., Kittaka, S., Nomura, T., Yonezawa, S. & Ishida, K. Evaluation of Spin-Triplet Superconductivity in Sr₂RuO₄. *J. Phys. Soc. Jpn.* **81**, 011009 (2012).
- Maeno, Y. *et al.* Superconductivity in a layered perovskite without copper. *Nature (London)* **372**, 532 (1994).
- Luke, G. M. *et al.* Time-reversal symmetry-breaking superconductivity in Sr₂RuO₄. *Nature (London)* **394**, 558–561 (1998).
- Ishida, K. *et al.* Spin-triplet superconductivity in Sr₂RuO₄ identified by ¹⁷O knight shift. *Nature (London)* **396**, 658 (1998).
- Nelson, K. D., Mao, Z. Q., Maeno, Y. & Liu, Y. Odd-parity superconductivity in Sr₂RuO₄. *Science* **306**, 1151 (2004).
- Xia, J., Maeno, Y., Beyersdorf, P. T., Fejer, M. M. & Kapitulnik, A. High resolution polar Kerr effect measurements of Sr₂RuO₄: Evidence for broken time-reversal symmetry in the superconducting state. *Phys. Rev. Lett.* **97**, 167002 (2006).
- Kidwingira, F., Strand, J. D., Harlingen, D. J. V. & Maeno, Y. Dynamical superconducting order parameter domains in Sr₂RuO₄. *Science* **314**, 1267 (2006).
- Nakamura, T. *et al.* Topological competition of superconductivity in Pb/Ru/Sr₂RuO₄ junctions. *Phys. Rev. B* **84**, 060512(R) (2011).
- Anwar, M. S. *et al.* Anomalous switching in Nb/Ru/Sr₂RuO₄ topological junctions by chiral domain wall motion. *Sci. Rep.* **3**, 2480 (2013).
- Anwar, M. S. *et al.* Multicomponent order parameter superconductivity of Sr₂RuO₄. *Phys. Rev. B* **95**, 224509 (2017).
- Hicks, C. W. *et al.* Limits on superconductivity-related magnetization in Sr₂RuO₄ and PrOs₄Sb₁₂ from scanning SQUID microscopy. *Phys. Rev. B* **81**, 214501 (2010).
- Yonezawa, S., Kajikawa, T. & Maeno, Y. First-order superconducting transition of Sr₂RuO₄. *Phys. Rev. Lett.* **110**, 077003 (2013).
- Hassinger, E. *et al.* Vertical line nodes in the superconducting gap structure of Sr₂RuO₄. *Phys. Rev. X* **7**, 011032 (2017).
- Pustogow, A. *et al.* Constraints on the superconducting order parameter in Sr₂RuO₄ from oxygen-17 nuclear magnetic resonance. *Nature(London)* **574**, 72 (2019).
- Ishida, K., Manago, M. & Maeno, Y. Reduction of the ¹⁷O Knight shift in the superconducting state and the heat-up effect by NMR pulses on Sr₂RuO₄. Preprint at, <https://arxiv.org/abs/1907.12236> (2019).
- Ng, K. K. & Sigrist, M. Anisotropy of the spin susceptibility in the normal state of Sr₂RuO₄. *J. Phys. Soc. Jpn.* **69**, 3764–3765 (2000).
- Ng, K. K. & Sigrist, M. The role of spin-orbit coupling for the superconducting state in Sr₂RuO₄. *Europhys. Lett.* **49**, 473 (2000).
- Annett, J. F., Litak, G., Györfy, B. L. & Wysokiński, K. I. Spin-orbit coupling and symmetry of the order parameter in strontium ruthenate. *Phys. Rev. B* **73**, 134501 (2006).
- Murakawa, H., Ishida, K., Kitagawa, K., Mao, Z. Q. & Maeno, Y. Measurement of the ¹⁰¹Ru-Knight shift of superconducting Sr₂RuO₄ in a parallel magnetic field. *Phys. Rev. Lett.* **93**, 167004 (2004).
- Annett, J. F., Györfy, B. L., Litak, G. & Wysokiński, K. I. Magnetic field induced rotation of the *d*-vector in the spin-triplet superconductor Sr₂RuO₄. *Phys. Rev. B* **78**, 054511 (2008).
- Anwar, M. S. *et al.* Ferromagnetic SrRuO₃ thin-film deposition on a spin-triplet superconductor Sr₂RuO₄ with a highly conducting interface. *Appl. Phys. Exp.* **8**, 019202 (2015).
- Anwar, M. S. *et al.* Direct penetration of spin-triplet superconductivity into a ferromagnet in Au/SrRuO₃/Sr₂RuO₄ junctions. *Nat. Commun.* **8**, 13220 (2016).
- Anwar, M. S. *et al.* Observation of superconducting gap spectra of long-range proximity effect in Au/SrTiO₃/SrRuO₃/Sr₂RuO₄ tunnel junctions. *Phys. Rev. B* **100**, 024516 (2019).
- Steppe, A. *et al.* Strong peak in T_c of Sr₂RuO₄ under uniaxial pressure. *Science* **355**, 133 (2017).
- Asulin, I., Yuli, O., Koren, G. & Millo, O. Evidence for crossed Andreev reflections in bilayers of (100) YBa₂Cu₃O_{7- δ} and the itinerant ferromagnet SrRuO₃. *Phys. Rev. B* **74**, 092501 (2006).
- Visani, C. *et al.* Equal-spin Andreev reflection and long-range coherent transport in high-temperature superconductor/half-metallic ferromagnet junctions. *Nat. Phys.* **8**, 539 (2012).
- Nomura, T. & Yamada, K. Detailed investigation of gap structure and specific heat in the *p*-wave superconductor Sr₂RuO₄. *J. Phys. Soc. Jpn.* **71**, 404 (2002).
- Rashba, E. I. Properties of semiconductors with an extremum loop. I. Cyclotron and combinational resonance in a magnetic field perpendicular to the plane of the loop. *Sov. Phys. Solid State* **2**, 1109 (1960).
- Lee, D. M. The extraordinary phases of liquid ³He. *Rev. Mod. Phys.* **69**, 645 (1997).
- Agterberg, D. F. Vortex Lattice Structures of Sr₂RuO₄. *Phys. Rev. Lett.* **80**, 5184 (1998).

49. Kaur, R. P., Agterberg, D. F. & Kusunose, H. Quasiclassical determination of the in-plane magnetic field phase diagram of superconducting Sr_2RuO_4 . *Phys. Rev. B* **72**, 144528 (2005).
50. Ishihara, M., Amano, Y., Ichioka, M. & Machida, K. Transverse magnetic field and chiral-nonchiral transition in vortex states for nearly B parallel to ab in chiral p -wave superconductors. *Phys. Rev. B* **87**, 224509 (2013).
51. Mao, Z. Q., Maeno, Y. & Fukazawa, H. Crystal growth of Sr_2RuO_4 . *Mater. Res. Bull.* **35**, 1813 (2000).

Acknowledgements

We are thankful to D. Manske, M. Couco and S. Arif for fruitful discussions. This work is supported by the JSPS KAKENHI projects Topological Quantum Phenomena (JP22103002 and JP25103721) and Topological Materials Science (JP15H05851, JP15K21717 and JP15H05852), JSPS KAKENHI 17H04848, as well as by JSPS-EPSRC core-to-core program “Oxide-Superspin (OSS)”. MSA is supported as an International Research Fellow of the JSPS. The work at IBS CCES is supported by the Institute of Basic Science (IBS) in Korea (Grants No. IBS-R009-D1). We acknowledge funding through the EPSRC (M.S.A. and J.W.A.R.) through International Network and Programme Grants (EP/P026311/1; EP/N017242/1).

Author contributions

M.S.A., S.Y. and Y.M. designed the experiments. S.R.L. grown the SRO113 thin films and preformed XDR experiments under supervision of T.W.N. R.I. and M.S.A. fabricated the devices. C.S. grew the single crystals. M.S.A. and M.K. prepared the SRO214 substrates and performed the measurements under supervision of Y.M. M.S.A., M.K. S.Y. and Y.M. wrote the manuscript. All authors contributed in data analysis, discussion and reviewing the manuscript.

Competing interests

The authors declare no competing interests.

Additional information

Correspondence and requests for materials should be addressed to M.S.A.

Reprints and permissions information is available at www.nature.com/reprints.

Publisher’s note Springer Nature remains neutral with regard to jurisdictional claims in published maps and institutional affiliations.



Open Access This article is licensed under a Creative Commons Attribution 4.0 International License, which permits use, sharing, adaptation, distribution and reproduction in any medium or format, as long as you give appropriate credit to the original author(s) and the source, provide a link to the Creative Commons license, and indicate if changes were made. The images or other third party material in this article are included in the article’s Creative Commons license, unless indicated otherwise in a credit line to the material. If material is not included in the article’s Creative Commons license and your intended use is not permitted by statutory regulation or exceeds the permitted use, you will need to obtain permission directly from the copyright holder. To view a copy of this license, visit <http://creativecommons.org/licenses/by/4.0/>.

© The Author(s) 2019