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PAPER

General boundary conditions for quasiclassical theory of superconductivity in the diffusive limit: application to strongly spin-polarized systems

M Eschrig[1,2], **A Cottet**[3], **W Belzig**[4] **and J Linder**[2]

1 Department of Physics, Royal Holloway, University of London, Egham, Surrey TW20 0EX, UK
2 Department of Physics, Norwegian University of Science and Technology, N-7491 Trondheim, Norway
3 Laboratoire Pierre Aigrain, Ecole Normale Supérieure-PSL Research University, CNRS, Université Pierre et Marie Curie-Sorbonne Universités, Université Paris Diderot-Sorbonne Paris Cité, 24 rue Lhomond, F-75231 Paris Cedex 05, France
4 Department of Physics, University of Konstanz, D-78457 Konstanz, Germany

E-mail: matthias.eschrig@rhul.ac.uk

Abstract

Boundary conditions in quasiclassical theory of superconductivity are of crucial importance for describing proximity effects in heterostructures between different materials. Although they have been derived for the ballistic case in full generality, corresponding boundary conditions for the diffusive limit, described by Usadel theory, have been lacking for interfaces involving strongly spin-polarized materials, e.g. half-metallic ferromagnets. Given the current intense research in the emerging field of superconducting spintronics, the formulation of appropriate boundary conditions for the Usadel theory of diffusive superconductors in contact with strongly spin-polarized ferromagnets for arbitrary transmission probability and arbitrary spin-dependent interface scattering phases has been a burning open question. Here we close this gap and derive the full boundary conditions for quasiclassical Green functions in the diffusive limit, valid for any value of spin polarization, transmission probability, and spin-mixing angles (spin-dependent scattering phase shifts). Our formulation allows also for complex spin textures across the interface and for channel off-diagonal scattering (a necessary ingredient when the numbers of channels on the two sides of the interface differ). As an example we derive expressions for the proximity effect in diffusive systems involving half-metallic ferromagnets. In a superconductor/half-metal/superconductor Josephson junction we find ϕ_0-junction behavior under certain interface conditions.

1. Introduction

Hybrid structures containing superconducting (S) and ferromagnetic (F) materials became a focus of nanoelectronic research because of their relevance for spintronics applications as well as their potential impact on fundamental research [1–3]. Examples of successful developments include the discoveries of the π-junction [4, 5] in S/F/S Josephson devices [6, 7], of odd-frequency superconductivity [8] in S/F heterostructures [9, 10], and of the indirect Josephson effect in S/half-metal/S junctions [11, 12]. Other recent topics of interest include the study of Majorana fermions at interfaces between superconductors and topological insulators [13] and at edges in superfluid ^{3}He [14, 15], and the appearance of pure spin supercurrents in topological superconductors [16], and in S/FI-F-FI devices as a result of geometric phases [17].

The central subject in many of these studies is to understand how in the case of a superconductor coupled to a ferromagnetic material superconducting correlations penetrate into the ferromagnet, and how magnetic correlations penetrate into the superconductor [18–23]. A powerful method to treat such problems is the quasiclassical theory of superconductivity developed by Larkin and Ovchinnikov and by Eilenberger [24, 25]. Within this theory [26–30] the quasiparticle motion is treated on a classical level, whereas the particle–hole and

the spin degrees of freedom are treated quantum mechanically. The transport equation, which is a first-order matrix differential equation for the quasiclassical propagator, must be supplemented by physical boundary conditions in order to obtain a unique solution.

Whereas for the full microscopic Green functions, i.e. the Gor'kov Green functions [31], such boundary conditions can be readily formulated (e.g. in terms of interface scattering matrices or in terms of transfer matrices), this is a considerably more difficult task for quasiclassical Green functions. In quasiclassical theory only the information about the envelope functions of Bloch waves is retained; information about the phases of the waves is missing. Such envelope amplitudes can show jumps at interfaces, and one complex task is to calculate these jumps without knowing the full microscopic Green functions near the interface. Correspondingly, there is a long history of deriving boundary conditions for quasiclassical propagators, both for the Eilenberger equations, and their diffusive limit, the Usadel equations [32].

For ballistic transport, described by the Eilenberger equations, such boundary conditions were first formulated for spin-inactive interfaces in pioneering work by Shelankov and by Zaitsev [34, 35], who showed the non-trivial fact that these jumps can be calculated using only the envelope functions. More general formulations were proposed subsequently [36–39], including a formulation in terms of interface scattering matrices by Millis, Rainer and Sauls [39]. All these formulations were implicit in terms of non-linear matrix equations, and problems arose in numerical implementations due to spurious (unphysical) additional solutions which must be eliminated. Progress was made with the help of Shelankov's projector formalism [40], allowing for explicit formulations of boundary conditions in both equilibrium [41–43] and non-equilibrium [42] situations. Further generalizations included spin-active interfaces, formulated for equilibrium [44] and for non-equilibrium [45], and interfaces with diffusive scattering characteristics [46]. An alternative formulation in terms of quantum mechanical *t*-matrices [47] proved also fruitful [11, 20, 48–51]. The latest formulation, in terms of interface scattering matrices, is able to include non-equilibrium phenomena, interfaces and materials with weak or strong spin polarization, multi-band systems, as well as disordered systems [52].

For the diffusive limit a set of second-order matrix differential equations was derived by Usadel [32]. In contrast to the ballistic case, where boundary conditions have been formulated for a wide set of applications, boundary conditions for the diffusive limit have been formulated so far only in certain limiting cases. The first formulation is by Kupriyanov and Lukichev, appropriate for the tunneling limit [53]. This was generalized to arbitrary transmission by Nazarov [54]. A major advance was done by Cottet *et al* in formulating boundary conditions for Usadel equations appropriate for spin-polarized interfaces [55]. These boundary conditions are valid in the limit of small transmission, spin polarization, and spin-dependent scattering phase shifts (this term is often used interchangeably with 'spin-mixing angles' [56]). Subsequent formulations allowed for arbitrary spin polarization, although being restricted to small transmission and spin-dependent scattering [57–59]. In [59] the authors present 'heuristically' deduced boundary conditions, which coincide with the ones used in [57, 58].

Here we not only present the full derivation of the specific boundary conditions used in [57–59], but go further and give a full solution of the problem. With this, the long-standing problem of how to generalize Nazarov's formula for arbitrary transmission probability [54] to the case of spin-polarized systems with arbitrary spin polarization and arbitrary spin dependent scattering phases is solved. Our boundary conditions are general enough to allow for non-equilibrium situations within Keldysh formalism, as well as for complex interface spin textures. We reproduce as limiting cases all previously known formulations.

2. Transport equations

The central quantity in quasiclassical theory of superconductivity [24, 25] is the quasiclassical Green function ('propagator') $\check{g}(\mathbf{p}_F, \mathbf{R}, E, t)$. It describes quasiparticles with energy E (measured from the Fermi level) and momentum $\mathbf{p}_F$ moving along classical trajectories with direction given by the Fermi velocity $\mathbf{v}_F(\mathbf{p}_F)$ in external potentials and self-consistent fields that are modulated by the slow spatial ($\mathbf{R}$) and time (t) coordinates [26–28]. The quasiclassical Green function is a functional of self-energies $\check{\Sigma}(\mathbf{p}_F, \mathbf{R}, E, t)$, which in general include molecular fields, the superconducting order parameter $\Delta(\mathbf{p}_F, \mathbf{R}, t)$, impurity scattering, and the external potentials. The quantum mechanical degrees of freedom of the quasiparticles show up in the matrix structure of the quasiclassical propagator and the self-energies. It is convenient to formulate the theory using 2×2 matrices in Keldysh space [60] (denoted by a 'check' accent), the elements of which in turn are 2×2 Nambu–Gor'kov matrices [31, 61] in particle–hole (denoted by a 'hat' accent) space. The structure of the propagators and self-energies in Keldysh-space is

$$\check{g} \;=\; \begin{pmatrix} \hat{g}^R & \hat{g}^K \\ 0 & \hat{g}^A \end{pmatrix}_{\mathrm{kel}}, \quad \check{\Sigma} \;=\; \begin{pmatrix} \hat{\Sigma}^R & \hat{\Sigma}^K \\ 0 & \hat{\Sigma}^A \end{pmatrix}_{\mathrm{kel}}, \tag{1a}$$

where the superscripts R, A and K refer to retarded, advanced and Keldysh components, respectively, and with the particle–hole space structure[5]

$$\hat{g}^{R,A} \;=\; \begin{pmatrix} g^{R,A} & f^{R,A} \\ \tilde{f}^{R,A} & \tilde{g}^{R,A} \end{pmatrix}_{\mathrm{ph}}, \quad \hat{g}^K \;=\; \begin{pmatrix} g^K & f^K \\ -\tilde{f}^K & -\tilde{g}^K \end{pmatrix}_{\mathrm{ph}} \tag{1b}$$

for Green functions, and

$$\hat{\Sigma}^{R,A} \;=\; \begin{pmatrix} \Sigma^{R,A} & \Delta^{R,A} \\ \tilde{\Delta}^{R,A} & \tilde{\Sigma}^{R,A} \end{pmatrix}_{\mathrm{ph}}, \quad \hat{\Sigma}^K \;=\; \begin{pmatrix} \Sigma^K & \Delta^K \\ -\tilde{\Delta}^K & -\tilde{\Sigma}^K \end{pmatrix}_{\mathrm{ph}} \tag{1c}$$

for self-energies. For spin-degenerate trajectories (i.e. in systems with weak or no spin-polarization) the elements of the 2×2 Nambu-Gor'kov matrices are 2×2 matrices in spin space, e.g. $g^R = g^R_{ab}$ with $a, b \in \{\uparrow, \downarrow\}$, and similarly for others. In strongly spin-polarized ferromagnets the elements of the 2×2 Nambu-Gor'kov matrices are spin-scalar (due to very fast spin-dephasing in a strong exchange field), and the system must be described within the preferred quantization direction given by the internal exchange field. The terms 'weak' and 'strong' refer to the spin-splitting of the energy bands being comparable to the superconducting gap or to the band width, respectively. In writing equations (1a)–(1c) we used general symmetries, which are accounted for by the 'tilde' operation,

$$\tilde{X}\left(\mathbf{p}_F, \mathbf{R}, E, t\right) \;=\; X\left(-\mathbf{p}_F, \mathbf{R}, -E, t\right)^{\star}. \tag{2}$$

Retarded (advanced) functions can be analytically continued into the upper (lower) complex energy half plane, in which case the relation is modified to $\tilde{X}(\mathbf{p}_F, \mathbf{R}, E, t) = X(-\mathbf{p}_F, \mathbf{R}, -E^{\star}, t)^{\star}$ with complex E.

The quasiclassical Green functions satisfy the Eilenberger–Larkin–Ovchinnikov transport equation and normalization condition

$$\left[E\check{\tau}_3 - \check{\Sigma}, \check{g}\right]_{\circ} + i\hbar\mathbf{v}_F \cdot \nabla\check{g} \;=\; \check{0}, \quad \check{g} \circ \check{g} \;=\; -\pi^2 \check{1}. \tag{3}$$

The non-commutative product $\circ$ combines matrix multiplication with a convolution over the internal energy-time variables in Wigner coordinate representation,

$$\left(\check{A} \circ \check{B}\right)(E, t) \;\equiv\; e^{\frac{i}{2}\left(\partial^A_E \partial^B_t - \partial^A_t \partial^B_E\right)} \check{A}(E, t)\check{B}(E, t), \tag{4}$$

and $\check{\tau}_3 = \hat{\tau}_3 \check{1}$, where $\hat{\tau}_3$ is a Pauli matrix in particle–hole space. Here and below, $[A, B]_{\circ} \equiv A \circ B - B \circ A$. The operation ∇ acts on the variable $\mathbf{R}$.

The functional dependence of the quasiclassical propagator on the self-energies is given in the form of self-consistency conditions. For instance, for a weak-coupling, s-wave order parameter, the condition reads

$$\hat{\Delta}(\mathbf{R}, t) \;=\; V_s \int_{-E_c}^{E_c} \frac{dE}{4\pi i} \left\langle N_F(\mathbf{p}_F)\hat{f}^K_s(\mathbf{p}_F, \mathbf{R}, E, t)\right\rangle_{\mathbf{p}_F}, \tag{5}$$

where V_s is the s-wave part of the singlet pairing interaction, N_F is the density of states per spin at the Fermi level, $\hat{f}^K_s$ is spin-singlet part of the the Keldysh component $\hat{f}^K$, and $\langle\rangle_{\mathbf{p}_F}$ denotes averaging over the Fermi surface. The cut-off energy E_c is to be eliminated in favor of the superconducting transition temperature in the usual manner.

When the quasiclassical Green function has been determined, physical quantities of interest can be calculated. For example, the current density at position $\mathbf{R}$ and time t reads (with $e < 0$ the electron charge)

$$\mathbf{j}(\mathbf{R}, t) \;=\; e \int_{-\infty}^{\infty} \frac{dE}{8\pi i} \, \mathrm{Tr}\left\langle N_F(\mathbf{p}_F)\mathbf{v}_F(\mathbf{p}_F)\hat{\tau}_3\hat{g}^K(\mathbf{p}_F, \mathbf{R}, E, t)\right\rangle_{\mathbf{p}_F}. \tag{6}$$

The symbol Tr denotes a trace over the 2×2 particle–hole space as well as over 2×2 spin space in the case of spin-degenerate trajectories.

In the dirty (diffusive) limit, strong scattering by non-magnetic impurities effectively averages the quasiclassical propagator over momentum directions. The Green function may then be expanded in the small parameter $k_B T_c \tau/\hbar$ (τ is the momentum relaxation time) following the standard procedure [32, 33]

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[5] For the definitions of all Green functions in this paper we use a basis of fermion field operators in Nambu $\otimes$ spin-space as
$\Psi(\mathbf{r}, t) = [\psi_\uparrow(\mathbf{r}, t), \psi_\downarrow(\mathbf{r}, t), \psi_\uparrow(\mathbf{r}, t)^{\dagger}, \psi_\downarrow(\mathbf{r}, t)^{\dagger}]^T$.

$$\check{g}\left(\mathbf{p}_F, \mathbf{R}, E, t\right) \approx \check{G}(\mathbf{R}, E, t) + \check{g}^{(1)}\left(\mathbf{p}_F, \mathbf{R}, E, t\right) \tag{7}$$

where the magnitude of $\check{g}^{(1)}$ is small compared to that of $\check{G}$. The impurity self-energy is related to an (in general anisotropic) lifetime function $\tau(\mathbf{p}_F', \mathbf{p}_F)$ [33]. Substitutings (7) into equation (3), multiplying with $N_F(\mathbf{p}_F') v_{F,j}(\mathbf{p}_F') \tau(\mathbf{p}_F', \mathbf{p}_F)$, averaging over momentum directions, considering that $\check{\Sigma}' \tau/\hbar$ is small, where $\check{\Sigma}'$ is the self-energy reduced by the contribution due to non-magnetic impurity scattering, and using $\check{G} \circ \check{G} = -\pi^2 \check{1}$ and $\check{G} \circ \check{g}^{(1)} + \check{g}^{(1)} \circ \check{G} = \check{0}$, one obtains (we suppress here the arguments $\mathbf{R}, E, t$)

$$\left\langle N_F(\mathbf{p}_F) v_{F,j}(\mathbf{p}_F) \check{g}^{(1)}(\mathbf{p}_F) \right\rangle_{\mathbf{p}_F} = N_F \sum_k \frac{D_{jk}}{i\pi} \check{G} \circ \nabla_k \check{G}, \tag{8}$$

where $N_F = \langle N_F(\mathbf{p}_F) \rangle_{\mathbf{p}_F}$ is the local density of states per spin at the Fermi level, $\nabla_k = \partial/\partial R_k$, the summation is over $k \in \{x, y, z\}$, and

$$D_{jk} = \frac{1}{N_F^2} \left\langle \left\langle N_F\left(\mathbf{p}_F'\right) v_{F,j}\left(\mathbf{p}_F'\right) \tau\left(\mathbf{p}_F', \mathbf{p}_F\right) v_{F,k}(\mathbf{p}_F) N_F(\mathbf{p}_F) \right\rangle_{\mathbf{p}_F} \right\rangle_{\mathbf{p}_F'} \tag{9}$$

is the diffusion constant tensor. For isotropic systems, $D_{jk} = D\delta_{jk}$. The Usadel Green function $\check{G}$ obeys the following transport equation and normalization condition, [32]

$$\left[E\check{\tau}_3 - \check{\Sigma}_0, \check{G} \right]_{\circ} + \sum_{jk} \frac{\hbar D_{jk}}{\pi} \nabla_j \left(\check{G} \circ \nabla_k \check{G} \right) = \check{0}, \quad \check{G} \circ \check{G} = -\pi^2 \check{1}, \tag{10}$$

where $\check{\Sigma}_0 = \langle N_F(\mathbf{p}_F) \check{\Sigma}'(\mathbf{p}_F) \rangle_{\mathbf{p}_F} / N_F$. The Usadel propagator $\check{G}$ is a functional of $\check{\Sigma}_0$.

The structures of $\check{G}$ and $\check{\Sigma}_0$ are the same as in equations (1*a*)–(1*c*) (with $\check{G}$ replacing $\check{g}$ and $\check{\Sigma}_0$ replacing $\check{\Sigma}_0$). Equation (2) is replaced by

$$\tilde{X}(\mathbf{R}, E, t) = X(\mathbf{R}, -E, t)^\star. \tag{11}$$

The current density for diffusive systems is obtained from equations (8) and (6), and is given by

$$j_i(\mathbf{R}, t) = -e \sum_k \int_{-\infty}^{\infty} \frac{\mathrm{d}E}{8\pi^2} \operatorname{Tr} N_F D_{ik} \hat{\tau}_3 \left[\check{G}(\mathbf{R}, E, t) \circ \nabla_k \check{G}(\mathbf{R}, E, t) \right]^K. \tag{12}$$

A vector potential $\mathbf{A}(\mathbf{R}, t)$ enters in a gauge invariant manner by replacing the spatial derivative operators in all expressions by (see e.g. [33, 62])

$$\nabla_i \hat{X} \to \hat{\partial}_i \circ \hat{X} \equiv \nabla_i \hat{X} - i \left[\frac{e}{\hbar} \hat{\tau}_3 A_i, \hat{X} \right]_{\circ}. \tag{13}$$

Finally, the case of a strongly spin-polarized itinerant ferromagnet with superconducting correlations (e.g. due to the proximity effect when in contact with a superconductor) can be treated by quasiclassical theory as well [11, 20, 50]. In this case, when the spin-splitting of the energy bands is comparable to the band width of the two spin bands, there exist two well-separated fully spin-polarized Fermi surfaces in the system, and the length scale associated with $\hbar/|\mathbf{p}_{F\uparrow} - \mathbf{p}_{F\downarrow}|$ is much shorter than the coherence length scale in the ferromagnet. Equal-spin correlations stay still coherent over long distance in such a system; $\uparrow\downarrow$ and $\downarrow\uparrow$ correlations are, however, incoherent and thus negligible within quasiclassical approximation. Fermi velocity, density of states, diffusion constant tensor, and coherence length all become spin-dependent. The quasiclassical propagator is then spin-scalar for each trajectory, with either all elements $\uparrow\uparrow$ or all elements $\downarrow\downarrow$ depending on the spin Fermi surface the trajectory corresponds to. Eilenberger equation and Usadel equation have the same form as before for each separate spin band. The spin-resolved current densities are given in the ballistic case by

$$\mathbf{j}_\uparrow = e \int_{-\infty}^{\infty} \frac{\mathrm{d}E}{8\pi i} \operatorname{Tr} \left\langle N_{F\uparrow} \mathbf{v}_{F\uparrow} \hat{\tau}_3 \hat{g}_{\uparrow\uparrow}^K \right\rangle_{\mathbf{p}_{F\uparrow}}, \tag{14}$$

and in the diffusive case by

$$j_{k\uparrow} = -e \sum_k \int_{-\infty}^{\infty} \frac{\mathrm{d}E}{8\pi^2} \operatorname{Tr} N_{F\uparrow} D_{\uparrow kj} \hat{\tau}_3 \left[\check{G}_{\uparrow\uparrow} \circ \nabla_j \check{G}_{\uparrow\uparrow} \right]^K, \tag{15}$$

and analogously for spin down.

For heterostructures, the above equations must be supplemented with boundary conditions at the interfaces. A practical formulation of boundary conditions for diffusive systems valid for arbitrary transmission and spin polarization is the goal of this paper.

3. Boundary conditions

3.1. Interface scattering matrix

We formulate boundary conditions at an interface in terms of the normal-state interface scattering matrix $\hat{\mathbf{S}}$ [63–65], connecting incoming with outgoing Bloch waves on either side of the interface with each other. We use the notation

$$\hat{\mathbf{S}} = \begin{pmatrix} \hat{\mathbf{S}}_{11} & \hat{\mathbf{S}}_{12} \\ \hat{\mathbf{S}}_{21} & -\hat{\mathbf{S}}_{22} \end{pmatrix}_{\times}, \tag{16}$$

where 1 and 2 refer to the two sides of the interface, and the subscript label $\times$ indicates that the 2×2 matrix structure refers to reflection and transmission amplitudes at an interface. The components $\hat{\mathbf{S}}_{ij}$ are matrices in particle–hole space as well as in scattering channel space (i.e. scattering channels for ballistic transport would be parameterized by the Fermi momenta of incoming and outgoing Bloch waves). Each element in 2×2 particle–hole space is in turn a matrix in combined spin and channel space, i.e. the number of incoming directions (assumed to be equal to the number of outgoing directions due to particle conservation) gives the dimension in channel space. The dimension in spin space is for spin-degenerate channels 2 and for spin-scalar channels 1.

If time-reversal symmetry is preserved, Kramers degeneracy requires that each element of the scattering matrix has a 2×2 spin (or more general: pseudo-spin) structure (as it connects doubly degenerate scattering channels on either side of the interface). For spin-polarized interfaces (e.g. ferromagnetic or with Rashba spin–orbit coupling) the scattering matrix is not spin-degenerate. However if the splitting of the spin-degeneracy is on the energy scale of the superconducting gap, it can be neglected within the precision of quasiclassical theory of superconductivity. On the other hand, if the lifting of the spin-degeneracy of energy bands is comparable to the Fermi energy, the degeneracy of the scattering channels must be lifted as well in order to achieve consistency within quasiclassical theory. For definiteness, we denote the dependence on the scattering channels by indices n, n':

$$\left[\hat{\mathbf{S}}_{\alpha\beta}\right]_{nn'}, \tag{17}$$

even for the ballistic case for which $[\hat{\mathbf{S}}_{\alpha\beta}]_{nn'} \equiv \hat{\mathbf{S}}_{\alpha\beta}(\mathbf{p}_{F,n}, \mathbf{k}_{F,n'})$.

As shown in appendices A and B, the scattering matrix for an interface can be written in polar decomposition in full generality as

$$\hat{\mathbf{S}} = \begin{pmatrix} \sqrt{1 - CC^{\dagger}} & C \\ C^{\dagger} & -\sqrt{1 - C^{\dagger}C} \end{pmatrix}_{\times} \begin{pmatrix} S & 0 \\ 0 & \check{S} \end{pmatrix}_{\times} \tag{18}$$

with unitary matrices S and $\check{S}$, and a transmission matrix C. All are matrices in particle–hole space, scattering channel space, and possibly (pseudo-)spin space. The above decomposition divides the scattering matrix into a Hermitian part and a unitary part. From this decomposition, we can define the auxiliary scattering matrix

$$\hat{\mathbf{S}}_0 = \begin{pmatrix} S & 0 \\ 0 & \check{S} \end{pmatrix}_{\times}, \tag{19}$$

which retains all the phase information during reflection on both sides of the interface, and has zero transmission components. The decomposition is uniquely defined when there are no zero-reflection singular values (we will assume here that a small non-zero reflection always takes place for each transmission channel; perfectly transmitting channels can always be treated separately as the corresponding boundary conditions are trivial). For the matrix C we introduce the parameterization

$$C = \left(1 + tt^{\dagger}\right)^{-1} 2t, \tag{20}$$

(see appendix C) which is uniquely defined when all singular values of t are in the interval $[0, 1]$ (which is required in order to ensure non-negative reflection singular values). We define for notational simplification 'hopping amplitude' matrices

$$\pi\tau_{12} = t\check{S}, \quad \pi\tau_{21} = t^{\dagger}S, \tag{21}$$

as well as unitary matrices

$$S_1 = S, \qquad S_2 = \check{S}. \tag{22}$$

In terms of those, obviously the relation

$$\tau_{\alpha\bar{\alpha}} \;=\; S_\alpha (\tau_{\bar{\alpha}\alpha})^\dagger S_{\bar{\alpha}} \tag{23}$$

holds, where $(\alpha, \bar{\alpha}) \in \{(1, 2), (2, 1)\}$, and the labels 1 and 2 refer to the respective sides of the interface. Here, and below, the Hermitian conjugate operation involves a transposition in channel indices. The particle–hole structures of the surface scattering matrix and the hopping amplitude are given by,

$$\hat{S}_\alpha \;=\; \begin{pmatrix} S_\alpha & 0 \\ 0 & \left(\tilde{S}_\alpha\right)^\dagger \end{pmatrix}_{\mathrm{ph}}, \qquad \hat{\tau}_{\alpha\bar{\alpha}} \;=\; \begin{pmatrix} \tau_{\alpha\bar{\alpha}} & 0 \\ 0 & \left(\tilde{\tau}_{\bar{\alpha}\alpha}\right)^\dagger \end{pmatrix}_{\mathrm{ph}}, \tag{24}$$

with

$$\left[\tilde{S}_\alpha\right]_{nn'} \;=\; \left[S_\alpha\right]^*_{\bar{n}\bar{n}'}, \quad \left[\tilde{\tau}_{\alpha\bar{\alpha}}\right]_{nn'} \;=\; \left[\tau_{\alpha\bar{\alpha}}\right]^*_{\bar{n}\bar{n}'}, \tag{25}$$

where $\bar{n}$ and $\bar{n}'$ denote mutually conjugated channels, e.g. defined by $\mathbf{p}_{F,\bar{n}'} \equiv -\mathbf{k}_{F,n'}$ and $\mathbf{k}_{F,\bar{n}} \equiv -\mathbf{p}_{F,n}$. Finally, the Keldysh structure of these quantities is

$$\check{S}_\alpha \;=\; \begin{pmatrix} \hat{S}_\alpha^R & 0 \\ 0 & \left(\hat{S}_\alpha^A\right)^\dagger \end{pmatrix}_{\mathrm{kel}} \equiv \begin{pmatrix} \hat{S}_\alpha & 0 \\ 0 & \hat{S}_\alpha \end{pmatrix}_{\mathrm{kel}}, \tag{26}$$

$$\check{\tau}_{\alpha\bar{\alpha}} \;=\; \begin{pmatrix} \hat{\tau}_{\alpha\bar{\alpha}}^R & 0 \\ 0 & \left(\hat{\tau}_{\bar{\alpha}\alpha}^A\right)^\dagger \end{pmatrix}_{\mathrm{kel}} \equiv \begin{pmatrix} \hat{\tau}_{\alpha\bar{\alpha}} & 0 \\ 0 & \hat{\tau}_{\alpha\bar{\alpha}} \end{pmatrix}_{\mathrm{kel}} \tag{27}$$

(the additional Hermitian conjugate in these equations is due to the fact that advanced Green functions have the roles of 'incoming' and 'outgoing' momentum directions interchanged compared to retarded Green functions; this is similar to the additional Hermitian conjugate appearing for hole components in particle–hole space). Thus, the Keldysh matrix structure for $\check{S}_\alpha$ and $\check{\tau}_{\alpha\bar{\alpha}}$ is trivial (proportional to the unit matrix). The full normal-state scattering matrix is diagonal in particle–hole and in Keldysh space, with reflection components

$$\check{\mathbf{S}}_{\alpha\alpha} \;=\; \left(1 + \pi^2 \check{\tau}_{\alpha\bar{\alpha}} \check{\tau}_{\alpha\bar{\alpha}}^\dagger\right)^{-1} \left(1 - \pi^2 \check{\tau}_{\alpha\bar{\alpha}} \check{\tau}_{\alpha\bar{\alpha}}^\dagger\right) \check{S}_\alpha, \tag{28}$$

and with transmission components

$$\check{\mathbf{S}}_{\alpha\bar{\alpha}} \;=\; \left(1 + \pi^2 \check{\tau}_{\alpha\bar{\alpha}} \check{\tau}_{\alpha\bar{\alpha}}^\dagger\right)^{-1} 2\pi \check{\tau}_{\alpha\bar{\alpha}}. \tag{29}$$

Note that $\check{\tau}_{\alpha\bar{\alpha}}$ connects incoming with outgoing Bloch waves per definition (as the scattering matrix does).

   We will formulate the theory such that all equations are valid on either side of the interface. This allows us to drop the indices α, $\bar{\alpha}$ for simplicity of notation by randomly choosing one side of the interface, and denoting quantities on the other side of the interface by underline. In particular, we will use
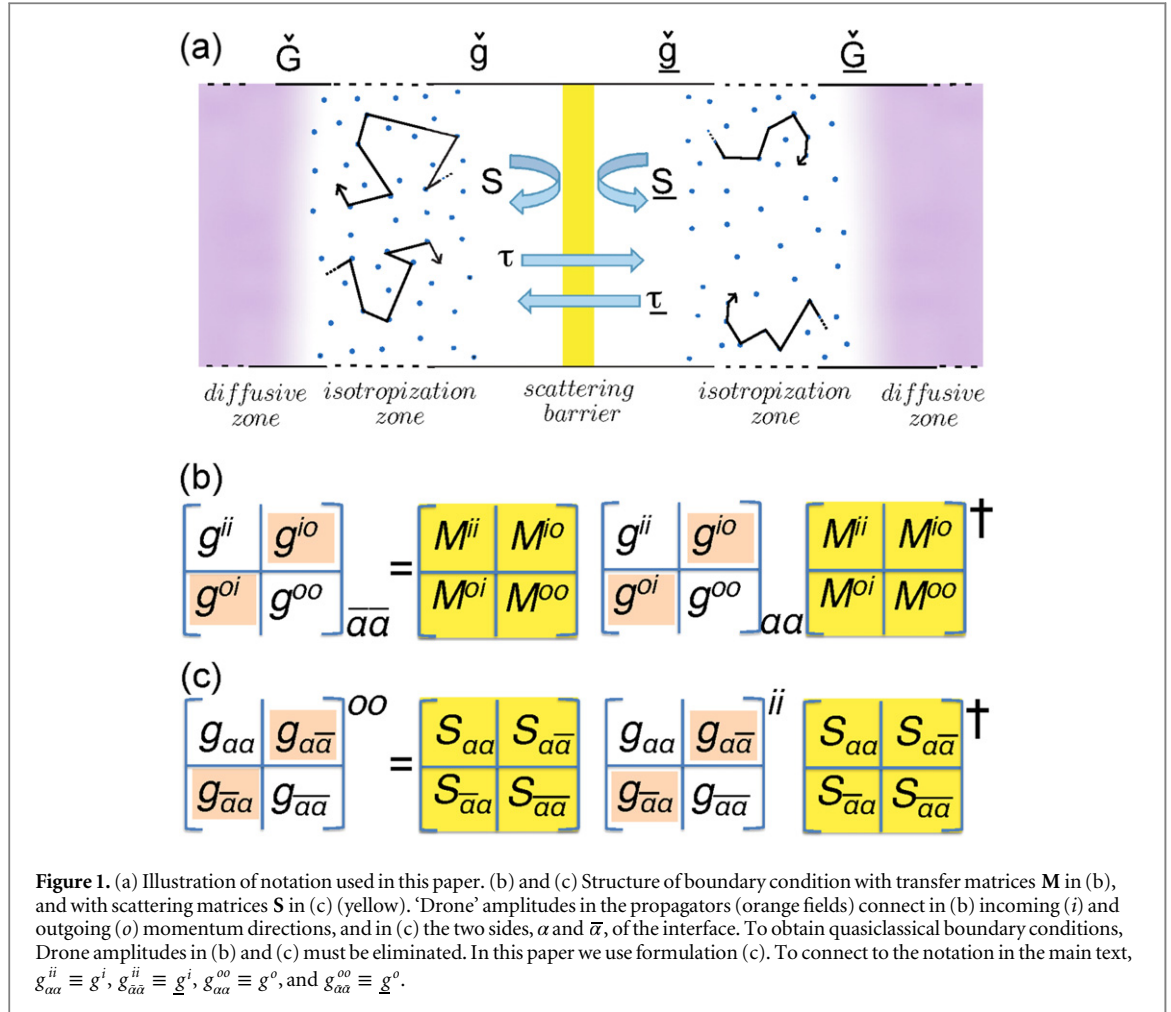
$$\check{S} \equiv \check{S}_\alpha, \quad \underline{\check{S}} \equiv \check{S}_{\bar{\alpha}}, \quad \check{\tau}_{\alpha\bar{\alpha}} \equiv \check{\tau}, \quad \check{\tau}_{\bar{\alpha}\alpha} \equiv \underline{\check{\tau}}$$

$$\check{g}_\alpha \equiv \check{g}, \quad \check{g}_{\bar{\alpha}} \equiv \underline{\check{g}}, \quad \check{G}_\alpha \equiv \check{G}, \quad \check{G}_{\bar{\alpha}} \equiv \underline{\check{G}}, \tag{30}$$

and so forth (see figure 1(a)). Also, from equation (23) we have $\check{\tau} \;=\; \check{S}\underline{\check{\tau}}^\dagger \underline{\check{S}}$.

3.2. General boundary conditions for diffusive systems

One main problem with boundary conditions for quasiclassical propagators is illustrated in figures 1(b) and (c). In previous treatments [39, 54, 55] the starting point was a transfer matrix description, see figure 1(b), which required the elimination of so-called 'drone amplitudes', which are propagators that mix incoming with outgoing directions. Here, we will employ a scattering matrix description, see figure 1(c), which, on the other hand, requires a similar elimination of Drone amplitudes, this time being propagators mixing the two sides of the interface. However, for an impenetrable interface this latter problem does not arise, a fact we will exploit.

   The strategy to derive the needed boundary conditions is to apply a three-step procedure. In the first step, the problem of an impenetrable interface with the auxiliary scattering matrix defined in equation (19) is solved on each side of the interface [11]. For this step, the ballistic solutions for the envelope functions for the Gor'kov propagators close to the interfaces should be expressed by the solutions $\check{G}$ of the Usadel equation. In a second step, these ballistic solutions (auxiliary propagators) are used in order to find the full ballistic solutions for finite transmission by utilizing a t-matrix technique [11, 20, 48, 50]. In the third and final step the matrix current will be derived from the ballistic solutions, which then enters the boundary conditions for the Usadel equations. We

Figure 1. (a) Illustration of notation used in this paper. (b) and (c) Structure of boundary condition with transfer matrices **M** in (b), and with scattering matrices **S** in (c) (yellow). 'Drone' amplitudes in the propagators (orange fields) connect in (b) incoming (*i*) and outgoing (*o*) momentum directions, and in (c) the two sides, α and $\bar{\alpha}$, of the interface. To obtain quasiclassical boundary conditions, Drone amplitudes in (b) and (c) must be eliminated. In this paper we use formulation (c). To connect to the notation in the main text, $g_{\alpha\alpha}^{ii} \equiv g^{i}$, $g_{\bar{\alpha}\bar{\alpha}}^{ii} \equiv \underline{g}^{i}$, $g_{\alpha\alpha}^{oo} \equiv g^{o}$, and $g_{\bar{\alpha}\bar{\alpha}}^{oo} \equiv \underline{g}^{o}$.

will present explicit solutions for all three steps, such that the procedure describes effectively boundary conditions for the solutions of Usadel equations on either side of the interface.

We use for the auxiliary propagators the notation $\check{g}_0^{o}$, $\check{g}_0^{i}$, $\underline{\check{g}}_0^{o}$ and $\underline{\check{g}}_0^{i}$, where the upper index denotes the direction of the Fermi velocity. *Incoming* momenta (index *i*) are those with a Fermi velocity pointing towards the interface, and *outgoing* momenta (index *o*) are those with a Fermi velocity pointing away from the interface.

3.2.1. Solution for impenetrable interface
We solve first for the auxiliary ballistic propagators fulfilling the impenetrable boundary conditions

$$\check{g}_0^{o} = \check{S}\,\check{g}_0^{i}\,\check{S}^{\dagger}, \qquad \underline{\check{g}}_0^{o} = \underline{\check{S}}\,\underline{\check{g}}_0^{i}\,\underline{\check{S}}^{\dagger}, \tag{31}$$

implying matrix multiplication in the combined (Keldysh) × (particle–hole) × (combined scattering-channel and spin) space. For diffusive banks, it is necessary to connect the ballistic propagators $\check{g}_0^{i,o}$ with the isotropic solutions of the Usadel equation, $\check{G}$. The ballistic propagators $\check{g}_0^{i,o}$ and $\underline{\check{g}}_0^{i,o}$, which characterize electronic correlations next to the scattering barrier, depend on the electronic momentum. However, in the diffusive case, impurity scattering leads to momentum isotropization away from the scattering barrier. This process occurs in isotropization zones with a thickness corresponding to a few times the inelastic mean-free path of the materials; see figure 1(a). This scale is itself much smaller than the scale on which the isotropic diffusive Green functions evolve in the bulk of the materials, in the framework of the Usadel equations. Indeed, the Usadel equations involve a superconducting coherence length, which is typically much larger than the elastic mean-free path. Therefore, in order to describe disordered hybrid structures with Usadel equations, suitable boundary conditions should be expressed in terms of the values of the isotropic Green functions $\check{G}$ and $\underline{\check{G}}$ right at the beginning of the isotropization zones. To obtain such boundary conditions from equation (31), it is necessary to express the propagators $\check{g}_0^{i,o}$ and $\underline{\check{g}}_0^{i,o}$ in terms of $\check{G}$ and $\underline{\check{G}}$. This can be done by studying the spatial dependence of the Gor'kov Green functions (or full Green functions without the quasiclassical approximation) in the isotropization zones (see [54, 55] for details). Using the fact that the dynamics of electrons is dominated by

impurity scattering in these zones, one can express the Gor'kov Green functions in terms of $\check{g}_0^{i,o}$, $\underline{\check{g}}_0^{i,o}$, $\check{G}$ and $\underline{\check{G}}$. Then, an elimination of unphysical solutions imposes the conditions [54]

$$\left(\check{G} - i\pi\check{1}\right) \circ \left(\check{g}_0^i + i\pi\check{1}\right) = \check{0}, \quad \left(\check{g}_0^i - i\pi\check{1}\right) \circ \left(\check{G} + i\pi\check{1}\right) = \check{0} \tag{32a}$$

$$\left(\check{G} + i\pi\check{1}\right) \circ \left(\check{g}_0^o - i\pi\check{1}\right) = \check{0}, \quad \left(\check{g}_0^o + i\pi\check{1}\right) \circ \left(\check{G} - i\pi\check{1}\right) = \check{0} \tag{32b}$$

and similarly for $\underline{\check{G}}$ and $\underline{\check{g}}_0^{i,o}$. From this one obtains the identity $\frac{1}{2}\left\{\check{g}_0^{i,o}, \check{G}\right\}_\circ = -\pi^2\check{1}$ for the anticommutator $\{\ldots\}$. This allows to solve after some straightforward algebra for $\check{g}_0^{i,o}$, using equation (31), and using the abbreviations

$$\check{G}' = \frac{1}{2\pi^2}\left(\check{S}^\dagger\check{G}\check{S} - \check{G}\right), \quad \check{G}'' = \frac{1}{2\pi^2}\left(\check{S}\check{G}\check{S}^\dagger - \check{G}\right), \tag{33}$$

(both are matrices depending via $\check{S}$ on the scattering channel index) leading to [55]

$$\check{g}_0^i - i\pi\check{1} = \left(1 - \check{G} \circ \check{G}'\right)^{-1} \circ \left(\check{G} - i\pi\check{1}\right), \tag{34a}$$

$$\check{g}_0^o + i\pi\check{1} = \left(1 - \check{G} \circ \check{G}''\right)^{-1} \circ \left(\check{G} + i\pi\check{1}\right) \tag{34b}$$

(here and below the inverse is defined with respect to the $\circ$-product), which, using identities like $\check{G}' \circ \check{G}' = -\frac{1}{2\pi^2}\{\check{G}', \check{G}\}_\circ$ (with $\{A, B\}_\circ \equiv A \circ B + B \circ A$), alternatively can be written also as

$$\check{g}_0^i + i\pi\check{1} = \left(\check{G} + i\pi\check{1}\right) \circ \left(1 - \check{G}' \circ \check{G}\right)^{-1}, \tag{34c}$$

$$\check{g}_0^o - i\pi\check{1} = \left(\check{G} - i\pi\check{1}\right) \circ \left(1 - \check{G}'' \circ \check{G}\right)^{-1}. \tag{34d}$$

Similar equations hold for $\underline{\check{G}}$ and $\underline{\check{g}}_0^{i,o}$ in terms of the scattering matrix $\underline{\check{S}}$. Introducing these solutions into equations (32a) and (32b) shows readily that the latter are fulfilled. We note that the relation $\check{g}_0^{i,o} \circ \check{g}_0^{i,o} = -\pi^2\check{1}$ follows from $\check{G} \circ \check{G} = -\pi^2\check{1}$ and $\check{S}\check{S}^\dagger = \check{S}^\dagger\check{S} = \check{1}$. It is also important to note that whereas $\check{G}$ is proportional to the unit matrix in channel space due to their isotropic nature [55], $\check{S}$, and consequently $\check{G}'$, $\check{G}''$, and $\check{g}_0^{i,o}$, are in general non-trivial matrices in channel space. Equations (34a) and (34b), or alternatively (34c) and (34d), together with equation (33) determine uniquely $\check{g}_0^{i,o}$ in terms of the diffusive Green function $\check{G}$. We can rewrite the difference $\check{g}_0^o - \check{g}_0^i$ in a more explicit manner, using the abbreviations $\check{\delta}' \equiv \check{G} \circ \check{G}'$ and $\check{\delta}'' \equiv \check{G}'' \circ \check{G}$, leading to

$$\check{g}_0^o - \check{g}_0^i = \left(\check{1} - \check{\delta}'\right)^{-1} \circ \left[\left(\check{G} - i\pi\check{1}\right) \circ \check{\delta}'' - \check{\delta}' \circ \left(\check{G} - i\pi\check{1}\right)\right] \circ \left(\check{1} - \check{\delta}''\right)^{-1}. \tag{35}$$

3.2.2. Solution for finite transmission

The second step follows [11, 20]. Once the auxiliary propagators are obtained, the full propagators can be obtained directly, without further solving the transport equation, in the following way. We solve t-matrix equations resulting from the transmission parameters $\check{\tau}$, for incoming and outgoing directions, which according to a procedure analogous to the one discussed in [47, 48] take the form,

$$\check{t}^i = \underline{\check{\tau}}^\dagger \underline{\check{g}}_0^o \underline{\check{\tau}} \circ \left(\check{1} + \check{g}_0^i \circ \check{t}^i\right), \quad \check{t}^o = \check{\tau} \underline{\check{g}}_0^i \check{\tau}^\dagger \circ \left(\check{1} + \check{g}_0^o \circ \check{t}^o\right). \tag{36}$$

Using the symmetry equation (23), the t-matrices for incoming and outgoing directions can be related through

$$\check{t}^o = \hat{S} \check{t}^i \hat{S}^\dagger. \tag{37}$$

Using the short notation

$$\check{g}_1^o \equiv \check{\tau} \underline{\check{g}}_0^i \check{\tau}^\dagger, \quad \check{g}_1^i \equiv \underline{\check{\tau}}^\dagger \underline{\check{g}}_0^o \underline{\check{\tau}}, \tag{38}$$

we solve formally equations (36) for $\check{t}^{i,o}$:

$$\check{t}^{i,o} = \left(1 - \check{g}_1^{i,o} \circ \check{g}_0^{i,o}\right)^{-1} \circ \check{g}_1^{i,o}. \tag{39}$$

The full propagators, fulfilling the desired boundary conditions at the interface, can now be easily calculated. For incoming and outgoing directions they are obtained from [11, 50]

$$\check{g}^i = \check{g}_0^i + \left(\check{g}_0^i + i\pi\check{1}\right) \circ \check{t}^i \circ \left(\check{g}_0^i - i\pi\check{1}\right), \tag{40a}$$

$$\check{g}^o = \check{g}_0^o + \left(\check{g}_0^o - i\pi\check{1}\right) \circ \check{t}^o \circ \left(\check{g}_0^o + i\pi\check{1}\right). \tag{40b}$$

Noticing that $\left(\check{g}_0^{i,o} + i\pi\check{1}\right) \circ \left(\check{g}_0^{i,o} - i\pi\check{1}\right) = \check{0}$, and $\left(\check{g}_0^{i,o} - i\pi\check{1}\right) \circ \left(\check{g}_0^{i,o} + i\pi\check{1}\right) = \check{0}$, as well as identities like $\check{g}_0^{i,o} \circ \left(\check{g}_0^{i,o} + i\pi\check{1}\right) = i\pi\check{1} \circ \left(\check{g}_0^{i,o} + i\pi\check{1}\right)$ etc, it is obvious that the normalization $\check{g}^{i,o} \circ \check{g}^{i,o} = -\pi^2\check{1}$ holds. Using the same identities, we obtain the alternative to equations (40a) and (40b) expressions

$$\check{g}^i = \check{g}_0^i + \left(\check{g}_0^i + i\pi\check{1}\right) \circ \left[\check{t}^i, \check{g}_0^i\right]_\circ = \check{g}_0^i - \left[\check{t}^i, \check{g}_0^i\right]_\circ \circ \left(\check{g}_0^i - i\pi\check{1}\right), \tag{40c}$$

$$\check{g}^o = \check{g}_0^o + \left(\check{g}_0^o - i\pi\check{1}\right) \circ \left[\check{t}^o, \check{g}_0^o\right]_\circ = \check{g}_0^o - \left[\check{t}^o, \check{g}_0^o\right]_\circ \circ \left(\check{g}_0^o + i\pi\check{1}\right). \tag{40d}$$

Equations (40a) and (40b), or alternatively, (40c) and (40d), in conjunction with equations (38) and (39), solve the problem of finding the ballistic solutions for finite transmission. We are now ready for the last step, to relate these solutions to the matrix current which enters in the expression for boundary conditions for $\check{G}$ and $\underline{\check{G}}$.

3.2.3. Matrix current and boundary conditions for diffusive propagators

We now turn to the third, final, step. As shown in [54, 55], the boundary conditions for quasiclassical isotropic Green functions can be obtained from the conservation of the matrix current $\mathcal{I}$ in the isotropization zones surrounding the scattering barrier. This quantity contains physical information on the flows of charge, spin and electron–hole coherence in a structure. We refer the reader to [54, 55] for the general definition of $\mathcal{I}$ in terms of the Gor'kov Green functions. Using this definition, one can verify that $\mathcal{I}$ is spatially conserved along the entire isotropization zones. Then, one can express $\mathcal{I}$ next to the scattering barrier in terms of the propagators $\check{g}^{i,o}$ and $\underline{\check{g}}^{i,o}$, and at the beginning of the isotropization zones in terms of $\check{G}$ and $\underline{\check{G}}$, see figure 1(a). The conservation of the matrix current provides an equality between the two expressions. Since $\check{g}^{i,o}$ can be expressed in terms of $\check{g}_0^{i,o}$ and $\underline{\check{g}}_0^{i,o}$, and these in terms of the $\check{G}$ and $\underline{\check{G}}$, this gives the desired boundary conditions. Following [50], after some straightforward algebra we obtain

$$\left[\check{t}^o, \check{g}_0^o\right]_\circ = \left(1 - \check{g}_1^o \circ \check{g}_0^o\right)^{-1} \left[\check{g}_1^o, \check{g}_0^o\right]_\circ \left(1 - \check{g}_0^o \circ \check{g}_1^o\right)^{-1}. \tag{41}$$

Using relations (31) and (37) above, we find

$$\check{g}^i = \check{S}^\dagger \left[\check{g}_0^o + \left(\check{g}_0^o + i\pi\check{1}\right) \circ \check{t}^o \circ \left(\check{g}_0^o - i\pi\check{1}\right)\right]\check{S}, \tag{42}$$

which allows to derive the following relation

$$\check{\mathcal{I}}' \equiv \check{g}^o - \check{S}\check{g}^i\check{S}^\dagger = -2\pi i\left[\check{t}^o, \check{g}_0^o\right]_\circ. \tag{43}$$

For calculating the charge current density in a given structure, it is sufficient to know $\check{\mathcal{I}}'$, because the matrices $\check{S}$ and $\check{S}^\dagger$ drop out of the trace as they commute with the $\hat{\tau}_3$ matrix in particle–hole space.

Finally we relate the obtained propagators $\check{g}^{i,o}$ to the matrix current $\mathcal{I}$,

$$\check{\mathcal{I}} \equiv \check{g}^o - \check{g}^i \equiv \check{\mathcal{I}}' + \check{\mathcal{I}}'' \tag{44}$$

with

$$\check{\mathcal{I}}'' \equiv \check{S}\check{g}^i\check{S}^\dagger - \check{g}^i. \tag{45}$$

We remind the reader here that $\check{\mathcal{I}}$ has a matrix structure in Keldysh space, in particle–hole space, and in combined scattering-channel and spin space. In terms of $\check{\mathcal{I}}$ the boundary condition results then from equation (8) and from the matrix current conservation in the isotropization regions [54]

$$\mathcal{G}_q \sum_{n=1}^{\mathcal{N}} \frac{\check{\mathcal{I}}_{nn}}{i\pi} = -\frac{\sigma\mathcal{A}}{\pi^2} \check{G} \circ \frac{d}{dz}\check{G}, \tag{46}$$

where z is the coordinate along the interface normal (*away from* the interface), n is a scattering channel index ($\mathcal{N}$ channels, spin-degenerate channels count as one), $\sigma = e^2 N_F D$ refers to the conductivity per spin, $\mathcal{A}$ is the surface area of the contact, and $\mathcal{G}_q$ is the quantum of conductance, $\mathcal{G}_q = e^2/h$. The number of scattering channels is expressed in terms of the projection of the Fermi surfaces on the contact plane, $A_{F,z}$, by $\mathcal{N} = A_{F,z}\mathcal{A}/(2\pi)^2$. For isotropic Fermi surfaces $A_{F,z} = \pi k_F^2$. In general,

$$\frac{1}{\mathcal{A}} \sum_{n=1}^{\mathcal{N}} \cdots = \int_{A_{F,z}} \frac{d^2 k_{||}}{(2\pi)^2} \cdots, \tag{47}$$

where $\hbar k_{||}$ is the momentum component parallel to the interface.

4. Special cases

4.1. Spin-scalar and channel-diagonal case

The transition to the diffusive Green functions is trivial for the case of $\hat{S} = \hat{1}$, as then $\check{g}_0^i = \check{g}_0^o = \check{G}$. If we start from equation (41) in conjunction with equation (38), we obtain in the case of a spin-scalar and channel-diagonal matrix $\hat{\tau}_{nn}$ with the notation $\check{G} = -i\pi\check{\mathbf{G}}$

$$\frac{2\sum_n \check{\mathcal{I}}_{nn}}{i\pi} = \sum_n \frac{4\mathcal{T}_n\left[\underline{\check{\mathbf{G}}}, \check{\mathbf{G}}\right]}{4 + \mathcal{T}_n\left(\left\{\underline{\check{\mathbf{G}}}, \check{\mathbf{G}}\right\} - 2\right)} = \frac{2\sigma\mathcal{A}}{\mathcal{G}_q}\check{\mathbf{G}} \circ \frac{\mathrm{d}}{\mathrm{d}z}\check{\mathbf{G}} \tag{48a}$$

with $\sigma = e^2 N_F D$ and

$$\mathcal{T}_n = \frac{4\pi^2|\tau_{nn}|^2}{\left(1 + \pi^2|\tau_{nn}|^2\right)^2}. \tag{48b}$$

This reproduces Nazarov's boundary condition [50, 54].

4.2. Case for interface between superconductor and ferromagnetic insulator

For the case of zero transmission, $\check{\tau} \equiv \check{0}$, we can find a closed solution if we assume that we can find a spin-diagonal basis for all reflection channels. For a channel-diagonal scattering matrix we write $\check{S}_{nn} = e^{i\varphi_n}e^{i\frac{\vartheta_n}{2}\check{\kappa}}$ with $\check{\kappa} = \mathrm{diag}\left\{\vec{m}\vec{\sigma}, \vec{m}\vec{\sigma}^\star\right\}$, where $\vec{m}^2 = 1$ (leading to $\check{\kappa}^2 = 1$). In this case we have $\check{g}^{i,o} = \check{g}_0^{i,o}$. We use equation (35), which straightforwardly leads to

$$\frac{2\sum_n \check{\mathcal{I}}_{nn}}{i\pi} = \sum_n \left[\check{1} - \frac{i\,\sin\vartheta_n}{2}\left(\check{\mathbf{G}}\check{\kappa}\check{\mathbf{G}} - \check{\kappa}\right) + \frac{\sin^2\frac{\vartheta_n}{2}}{2}\left(\check{\mathbf{G}}\check{\kappa}\check{\mathbf{G}}\check{\kappa} - \check{1}\right)\right]^{-1}$$
$$\times \left\{-i\sin\vartheta_n\left[\check{\kappa}, \check{\mathbf{G}}\right] + \sin^2\frac{\vartheta_n}{2}\left[\check{\kappa}\check{\mathbf{G}}\check{\kappa}, \check{\mathbf{G}}\right]\right\}$$
$$\times \left[\check{1} - \frac{i\,\sin\vartheta_n}{2}\left(\check{\mathbf{G}}\check{\kappa}\check{\mathbf{G}} - \check{\kappa}\right) + \frac{\sin^2\frac{\vartheta_n}{2}}{2}\left(\check{\kappa}\check{\mathbf{G}}\check{\kappa}\check{\mathbf{G}} - \check{1}\right)\right]^{-1} \tag{49}$$

(where we recall that $\check{\mathbf{G}}^2 = \check{1}$). Note that φ_n drops out, and only the spin mixing angle ϑ_n matters. Equation (49) generalizes the results of [55] to arbitrary spin-dependent reflection phases. Further below we will give a physical interpretation of the leading order terms arising in an expansion for small ϑ_n.

4.3. Exact series expansions

We now provide explicit series expansions for all quantities which will be useful for deriving formulas for various limiting cases. We start with writing the scattering matrix as $\check{S} = e^{i\check{K}}$ with hermitian $\check{K}$ due to unitarity of $\check{S}$, i.e. $\check{K} = \check{K}^\dagger$. Then we use an expansion formula for Lie brackets in order to obtain the series expansion

$$\check{S}^\dagger\check{G}\check{S} = e^{-i\check{K}}\check{G}e^{i\check{K}} = \sum_{m=0}^{\infty}\frac{(-i)^m}{m!}\left[\check{K}^m, \check{G}\right] \tag{50}$$

with the definitions $\left[\check{K}^m, \check{G}\right] = \left[\check{K}, \left[\check{K}^{m-1}, \check{G}\right]\right]$ and $\left[\check{K}^0, \check{G}\right] = \check{G}$. With this we obtain from equation (33)

$$\check{G}' = \frac{1}{2\pi^2}\sum_{m=1}^{\infty}\frac{(-i)^m}{m!}\left[\check{K}^m, \check{G}\right], \qquad \check{G}'' = \frac{1}{2\pi^2}\sum_{m=1}^{\infty}\frac{i^m}{m!}\left[\check{K}^m, \check{G}\right], \tag{51}$$

which are very useful if $\check{K}$ has a small pre-factor. Note also the identity $\check{G} \circ \left[\check{K}, \check{G}\right] \circ \check{G} = \pi^2\left[\check{K}, \check{G}\right]$. Furthermore, from equations (34c) and (34d) we find

$$\check{g}_0^i = \check{G} + \left(\check{G} + i\pi\check{1}\right) \circ \sum_{l=1}^{\infty}(\check{G}' \circ \check{G})^l \tag{52a}$$

$$\check{g}_0^{\,o} \;=\; \check{G} + \left(\check{G} - i\pi\check{1}\right)\circ \sum_{l=1}^{\infty} (\check{G}'' \circ \check{G})^l. \tag{52b}$$

From equation (41), and using equations (31), (37), we derive

$$\left[\check{t}^{\,o}, \check{g}_0^{\,o}\right]_{\circ} \;=\; \sum_{k,n=0}^{\infty} \left(\check{g}_1^{\,o}\circ\check{g}_0^{\,o}\right)^k \circ \left[\check{g}_1^{\,o}, \check{g}_0^{\,o}\right]_{\circ} \circ \left(\check{g}_0^{\,o}\circ\check{g}_1^{\,o}\right)^n, \tag{53a}$$

$$\left[\check{t}^{\,i}, \check{g}_0^{\,i}\right]_{\circ} \;=\; \sum_{k,n=0}^{\infty} \left(\check{g}_1^{\,i}\circ\check{g}_0^{\,i}\right)^k \circ \left[\check{g}_1^{\,i}, \check{g}_0^{\,i}\right]_{\circ} \circ \left(\check{g}_0^{\,i}\circ\check{g}_1^{\,i}\right)^n, \tag{53b}$$

which is useful if the transmission amplitudes $\check{\tau}$ entering into $\check{g}_1^{\,i,o}$ are small. Finally, we obtain from equations (43) and (45)

$$\check{I}' \;=\; -2\pi i\left[\check{t}^{\,o}, \check{g}_0^{\,o}\right]_{\circ}, \qquad \check{I}'' \;=\; \sum_{m=1}^{\infty} \frac{i^m}{m!}\left[\check{K}^{\,m}_{\cdot\cdot}, \check{g}^{\,i}\right]. \tag{54}$$

Here, $\check{g}^{\,i}$ is obtained from

$$\check{g}^{\,i} + i\pi\check{1} \;=\; \left(\check{G} + i\pi\check{1}\right)\circ \sum_{l=0}^{\infty}(\check{G}'\circ\check{G})^l \circ \left(\check{1} + \left[\check{t}^{\,i}, \check{g}_0^{\,i}\right]_{\circ}\right). \tag{55}$$

4.4. Boundary condition for spin-polarized surface to third order in spin-mixing angles

We first treat the case when $\check{t}^{\,i,o} \equiv \check{0}$, for example the case where one side of the junction is a ferromagnetic insulator (FI). Then

$$\check{I} \;=\; \sum_{m=1}^{\infty} \frac{i^m}{m!}\left[\check{K}^{\,m}_{\cdot\cdot}, \check{G}\right] + \sum_{m,l=1}^{\infty} \frac{i^m}{m!}\left[\check{K}^{\,m}_{\cdot\cdot}, \left(\check{G} + i\pi\check{1}\right)\circ(\check{G}'\circ\check{G})^l\right]. \tag{56}$$

To third order we have $\check{I} \;=\; \check{I}^{(1)} + \check{I}^{(2)} + \check{I}^{(3)}$, and the derivation in appendix D leads to

$$\check{I}^{(1)} \;=\; i\left[\check{K}, \check{G}\right], \qquad \check{I}^{(2)} \;=\; -\frac{i}{2\pi}\left[\check{K}\check{G}\check{K}, \check{G}\right]_{\circ}. \tag{57a}$$

$$\check{I}^{(3)} \;=\; -\frac{i}{24}\left[\check{K}^{\,3}_{\cdot}, \check{G}\right] - \frac{i}{8\pi^2}\left[\check{K}, \check{G}\circ\left[\check{K}^{\,2}_{\cdot}, \check{G}\right]\circ\check{G}\right]. \tag{57b}$$

For the special case of channel diagonal $\check{K}_{nn} \;=\; \frac{\vartheta_n}{2}\check{\kappa}$ with $\check{\kappa}^2 \;=\; \check{1}$, which follows also from directly expanding equation (49), we reproduce the results from [55] ($\check{G} \;=\; -i\pi\check{\mathbf{G}}$),

$$\frac{2\sum_n \check{I}^{(1)}_{nn}}{i\pi} \;=\; -i\left(\sum_n \vartheta_n\right)\left[\check{\kappa}, \check{\mathbf{G}}\right], \qquad \frac{2\sum_n \check{I}^{(2)}_{nn}}{i\pi} \;=\; \frac{\sum_n \vartheta_n^2}{4}\left[\check{\kappa}\check{\mathbf{G}}\check{\kappa}, \check{\mathbf{G}}\right]_{\circ}. \tag{58a}$$

$$\frac{2\sum_n \check{I}^{(3)}_{nn}}{i\pi} \;=\; -i\frac{\sum_n \vartheta_n^3}{16}\left(\frac{1}{3}\left[\check{\kappa}, \check{\mathbf{G}}\right] - \left[\check{\kappa}\check{\mathbf{G}}\check{\kappa}\circ\check{\mathbf{G}}\check{\kappa}, \check{\mathbf{G}}\right]_{\circ}\right). \tag{58b}$$

Note that the first order term $\sim[\check{\kappa}, \check{\mathbf{G}}]$ accounts for the effective exchange field induced inside the superconductor by the spin-mixing, whereas the term $\sim[\check{\kappa}\check{\mathbf{G}}\check{\kappa}, \check{\mathbf{G}}]$ produces a pair breaking effect similar to that of paramagnetic impurities [66]. This second term occurs only at second order in ϑ_n because it requires multiple scattering at the S/FI interface, which together with random scattering in the diffusive superconductor leads to a magnetic disorder effect.

4.5. Boundary condition for spin-polarized interface to second order in spin-mixing angles and transmission probability

We now allow for finite transmission, and concentrate on the matrix current to second order in the quantities $\check{K}$, $\underline{\check{K}}$, and $\check{g}_1^{\,i,o}$. We need to take care of the scattering phases during transmission events. For this, we define

$$\check{\tau} \;=\; \check{S}^{\frac{1}{2}}\check{\tau}_0\underline{\check{S}}^{\frac{1}{2}}, \qquad \underline{\check{\tau}} \;=\; \underline{\check{S}}^{\frac{1}{2}}\underline{\check{\tau}}_0\check{S}^{\frac{1}{2}}. \tag{59}$$

We note that equation (23), or $\check{\tau} \;=\; \check{S}\underline{\check{\tau}}^\dagger\underline{\check{S}}$, results into

$$\check{\tau}_0 \;=\; \underline{\check{\tau}}_0^\dagger. \tag{60}$$

Thus, the $\check{\tau}_0$ and $\underline{\check{\tau}}_0$ are the appropriate transmission amplitudes, with transmission spin-mixing phases removed. We further define

$$\check{G}_1 \equiv \tau_0 \underline{\check{G}} \tau_0^{\dagger}. \tag{61}$$

We expand $\check{\tau}$ up to first order in $\check{K}$ and $\underline{\check{K}}$,

$$\check{\tau} \;=\; \check{\tau}_0 + \frac{\mathrm{i}}{2} \left(\check{K} \check{\tau}_0 + \check{\tau}_0 \underline{\check{K}} \right) + \ldots, \tag{62}$$

and obtain $\check{\mathcal{I}} \;=\; \check{\mathcal{I}}^{(1)} + \check{\mathcal{I}}^{(2)}$ from a systematic expansion to second order in $\check{K}$, $\underline{\check{K}}$, and $\check{G}_1$, as shown in appendix E, leading to one of the main results of this paper:

$$\check{\mathcal{I}}^{(1)} \;=\; -2\pi\mathrm{i} \left[\check{G}_1, \check{G} \right]_{\circ} + \mathrm{i} \left[\check{K}, \check{G} \right], \tag{63a}$$

$$\begin{aligned}
\check{\mathcal{I}}^{(2)} \;=\;\; & -2\pi\mathrm{i} \left[\check{G}_1 \circ \check{G} \circ \check{G}_1, \check{G} \right]_{\circ} - \frac{\mathrm{i}}{2\pi} \left[\check{K} \check{G} \check{K}, \check{G} \right]_{\circ} \\
& + \mathrm{i} \left[\check{G}_1 \circ \check{G} \check{K} + \check{K} \check{G} \circ \check{G}_1 + \check{\tau}_0 \underline{\check{G}} \circ \left[\underline{\check{K}}, \underline{\check{G}} \right] \check{\tau}_0^{\dagger}, \check{G} \right]_{\circ}.
\end{aligned} \tag{63b}$$

These relations generalize the results of [55] for the case of arbitrary spin polarization, and are valid even when $\check{K}$, $\underline{\check{K}}$ and τ have different spin quantization axes, i.e. cannot be diagonalized simultaneously.

Using the notation $\check{G} \;=\; -\mathrm{i}\pi\check{\mathbf{G}}$ and $2\pi\check{\tau}_0 \;=\; \check{T}$, we can rewrite the result in leading order in the quantities $\check{K}$, $\underline{\check{K}}$, and the transmission probability ($\sim \check{T}\check{T}^{\dagger}$) as

$$\frac{2\check{\mathcal{I}}^{(1)}}{\mathrm{i}\pi} \;=\; \left[\check{T} \underline{\check{\mathbf{G}}} \check{T}^{\dagger} - 2\mathrm{i}\check{K}, \check{\mathbf{G}} \right]_{\circ}, \tag{64a}$$

and for the next-to-leading order

$$\begin{aligned}
\frac{2\check{\mathcal{I}}^{(2)}}{\mathrm{i}\pi} \;=\;\; & -\frac{1}{4} \left[\check{T} \underline{\check{\mathbf{G}}} \check{T}^{\dagger} \circ \check{\mathbf{G}} \circ \check{T} \underline{\check{\mathbf{G}}} \check{T}^{\dagger}, \check{\mathbf{G}} \right]_{\circ} + \left[\check{K} \check{\mathbf{G}} \check{K}, \check{\mathbf{G}} \right]_{\circ} \\
& + \frac{\mathrm{i}}{2} \left[\check{T} \underline{\check{\mathbf{G}}} \check{T}^{\dagger} \circ \check{\mathbf{G}} \check{K} + \check{K} \check{\mathbf{G}} \circ \check{T} \underline{\check{\mathbf{G}}} \check{T}^{\dagger} + \check{T}\underline{\check{\mathbf{G}}} \circ \left[\underline{\check{K}}, \underline{\check{\mathbf{G}}} \right] \check{T}^{\dagger}, \check{\mathbf{G}} \right]_{\circ}.
\end{aligned} \tag{64b}$$

These equations are still fully general with respect to the magnetic (spin) structure, and allow for channel off-diagonal scattering as well as different numbers of channels on the two sides of the interface. Note that $\check{T}$, $\check{K}$, and $\underline{\check{K}}$ are matrices in channel space, whereas $\check{\mathbf{G}}$ and $\underline{\check{\mathbf{G}}}$ are proportional to the unit matrix in channel space. Whereas $\check{K}$, and $\underline{\check{K}}$ are square matrices, $\check{T}$ in general can be a rectangular matrix (when the number of channels on the two sides of the interface differ).

4.6. Boundary conditions for channel-independent spin quantization direction

As an application, we assume next that each of the quantities $\check{K}$, $\underline{\check{K}}$, and $\check{\tau}_0$ can be spin-diagonalized simultaneously for all channels, with spin quantization directions $\vec{m}'$, $\underline{\vec{m}}'$, and $\vec{m}$ for $\check{K}$, $\underline{\check{K}}$, or $\check{\tau}_0$, respectively. We also use that $\check{\mathbf{G}}$ and $\underline{\check{\mathbf{G}}}$ are proportional to the unit matrix in channel space, as they are isotropic [55], and we assume that the number of channels on both sides of the interface are equal. We define

$$\mathbb{T}_{0,nl} \,\check{1} + \mathbb{T}_{1,nl} \, \vec{m} \cdot \vec{\sigma} \;=\; \check{T}_{nl}, \tag{65a}$$

$$\varphi_{nn'} \,\check{1} + \frac{1}{2} \vartheta_{nn'} \, \vec{m}' \cdot \vec{\sigma} \;=\; \check{K}_{nn'}, \qquad \underline{\varphi}_{ll'} \,\check{1} + \frac{1}{2} \underline{\vartheta}_{ll'} \, \underline{\vec{m}}' \cdot \vec{\sigma} \;=\; \underline{\check{K}}_{ll'}, \tag{65b}$$

$$\vec{\check{\sigma}} \;=\; \vec{\sigma}\check{1}, \qquad \vec{\check{\sigma}} \;=\; \begin{pmatrix} \vec{\sigma} & 0 \\ 0 & \vec{\sigma}^{\star} \end{pmatrix}_{\mathrm{ph}}, \quad \check{\kappa} \equiv \vec{m} \cdot \vec{\check{\sigma}}, \quad \check{\kappa}' \equiv \vec{m}' \cdot \vec{\check{\sigma}}, \quad \underline{\check{\kappa}}' \equiv \underline{\vec{m}}' \cdot \vec{\check{\sigma}} \tag{65c}$$

with $\vec{m}^2 \;=\; (\vec{m}')^2 \;=\; (\underline{\vec{m}}')^2 \;=\; 1$, i.e. $\check{\kappa}^2 \;=\; (\check{\kappa}')^2 \;=\; (\underline{\check{\kappa}}')^2 \;=\; \check{1}$, and introduce the transmission probability $\mathcal{T}_{nl}$ and the spin polarization $\mathcal{P}_{nl}$ as

$$\mathcal{T}_{nl} \left(\check{1} + \mathcal{P}_{nl} \vec{m}\vec{\check{\sigma}} \right) \;=\; \check{T}_{nl} \left[\check{T}_{nl} \right]^{\dagger}. \tag{66}$$

We write for $\mathbb{T}_{0,nl}$ and $\mathbb{T}_{1,nl}$, allowing for some spin-scalar phases ψ_{nl},

$$\mathbb{T}_{0,nl}^2 \;=\; \frac{\mathcal{T}_{nl}}{2} \left[1 + \sqrt{1 - \mathcal{P}_{nl}^2} \right] \mathrm{e}^{2\mathrm{i}\psi_{nl}}, \qquad \mathbb{T}_{1,nl}^2 \;=\; \frac{\mathcal{T}_{nl}}{2} \left[1 - \sqrt{1 - \mathcal{P}_{nl}^2} \right] \mathrm{e}^{2\mathrm{i}\psi_{nl}}. \tag{67}$$

We will average over all spin-scalar phases ψ_{nl} of the transmission amplitudes as there are usually many scattering channels in an area comparable with the superconducting coherence length squared. This filters out all the terms in equations (64a) and (64b) where these scalar scattering phases cancel.

For a magnetic system, in linear order in $\mathcal{T}_{nl}$ and $\vartheta_{nn'}$ we obtain

$$I^{(1)} \equiv \frac{2\mathcal{G}_q \sum_n \check{\mathcal{I}}_{nn}^{(1)}}{\mathrm{i}\pi} = \mathcal{G}_q \sum_{nl}\Big[\big(\mathbb{T}_{0,nl}\check{\mathbb{1}} + \mathbb{T}_{1,nl}\check{\kappa}\big)\underline{\check{\mathbf{G}}}\big(\mathbb{T}_{0,nl}^{\star}\check{\mathbb{1}} + \mathbb{T}_{1,nl}^{\star}\check{\kappa}\big), \check{\mathbf{G}}\Big]$$
$$- \mathcal{G}_q \sum_n \mathrm{i}\vartheta_{nn}\big[\check{\kappa}', \check{\mathbf{G}}\big], \tag{68}$$

where $\mathcal{G}_q = e^2/h$ is the conductance quantum. After multiplying out we obtain the set of boundary conditions

$$2I^{(1)} = \Big[\mathcal{G}^0\underline{\check{\mathbf{G}}} + \mathcal{G}^{\mathrm{MR}}\big\{\check{\kappa}, \underline{\check{\mathbf{G}}}\big\} + \mathcal{G}^1\check{\kappa}\underline{\check{\mathbf{G}}}\check{\kappa} - \mathrm{i}\mathcal{G}^{\phi}\check{\kappa}', \check{\mathbf{G}}\Big]_{\circ} \tag{69a}$$

with

$$\mathcal{G}^0 = \mathcal{G}_q \sum_{nl} \mathcal{T}_{nl}\Big(1 + \sqrt{1 - \mathcal{P}_{nl}^2}\Big) \tag{69b}$$

$$\mathcal{G}^1 = \mathcal{G}_q \sum_{nl} \mathcal{T}_{nl}\Big(1 - \sqrt{1 - \mathcal{P}_{nl}^2}\Big) \tag{69c}$$

$$\mathcal{G}^{\mathrm{MR}} = \mathcal{G}_q \sum_{nl} \mathcal{T}_{nl}\mathcal{P}_{nl}, \quad \mathcal{G}^{\phi} = 2\mathcal{G}_q \sum_n \vartheta_{nn} \tag{69d}$$

For $\kappa = \kappa'$ and the assumption of a channel-diagonal scattering matrix ($n = l$) this also provides the derivation of the boundary conditions used for [57]. We now proceed to the second-order terms:

$$2I^{(2)} = -2I_4 + \mathcal{G}_2^{\phi}\big[\check{\kappa}'\check{\mathbf{G}}\check{\kappa}', \check{\mathbf{G}}\big]_{\circ} + \mathrm{i}\big[\check{\mathbf{M}}_{\chi,\underline{\chi}}^0 + \check{\mathbf{M}}_{\chi,\underline{\chi}}^1 + \check{\mathbf{M}}_{\chi,\underline{\chi}}^{\mathrm{MR}}, \check{\mathbf{G}}\big]_{\circ}$$

$$\check{\mathbf{M}}_{\chi,\underline{\chi}}^0 = \mathcal{G}_{\chi}^0 \big(\underline{\check{\mathbf{G}}} \circ \check{\mathbf{G}}\check{\kappa}' + \check{\kappa}'\check{\mathbf{G}} \circ \underline{\check{\mathbf{G}}}\big) + \mathcal{G}_{\underline{\chi}}^0 \,\underline{\check{\mathbf{G}}} \circ \big[\underline{\check{\kappa}}', \underline{\check{\mathbf{G}}}\big]$$

$$\check{\mathbf{M}}_{\chi,\underline{\chi}}^1 = \mathcal{G}_{\chi}^1 \big(\check{\kappa}\underline{\check{\mathbf{G}}}\check{\kappa} \circ \check{\mathbf{G}}\check{\kappa}' + \check{\kappa}'\check{\mathbf{G}} \circ \check{\kappa}\underline{\check{\mathbf{G}}}\check{\kappa}\big) + \mathcal{G}_{\underline{\chi}}^1 \,\check{\kappa}\underline{\check{\mathbf{G}}} \circ \big[\underline{\check{\kappa}}', \underline{\check{\mathbf{G}}}\big]\check{\kappa}$$

$$\check{\mathbf{M}}_{\chi,\underline{\chi}}^{\mathrm{MR}} = \mathcal{G}_{\chi}^{\mathrm{MR}}\big(\big\{\check{\kappa}, \underline{\check{\mathbf{G}}}\big\} \circ \check{\mathbf{G}}\check{\kappa}' + \check{\kappa}'\check{\mathbf{G}} \circ \big\{\check{\kappa}, \underline{\check{\mathbf{G}}}\big\}\big) + \mathcal{G}_{\underline{\chi}}^{\mathrm{MR}}\big\{\check{\kappa}, \underline{\check{\mathbf{G}}} \circ \big[\underline{\check{\kappa}}', \underline{\check{\mathbf{G}}}\big]\big\} \tag{70a}$$

where I_4 denotes a cumbersome expression in fourth order of the transmission amplitudes, which we do not write down here explicitly (see appendix F). We have used the abbreviations

$$\mathcal{G}_{\chi}^0 = \frac{1}{4}\mathcal{G}_q \sum_{nl} \vartheta_{nn}\mathcal{T}_{nl}\Big(1 + \sqrt{1 - \mathcal{P}_{nl}^2}\Big) \tag{70b}$$

$$\mathcal{G}_{\chi}^1 = \frac{1}{4}\mathcal{G}_q \sum_{nl} \vartheta_{nn}\mathcal{T}_{nl}\Big(1 - \sqrt{1 - \mathcal{P}_{nl}^2}\Big) \tag{70c}$$
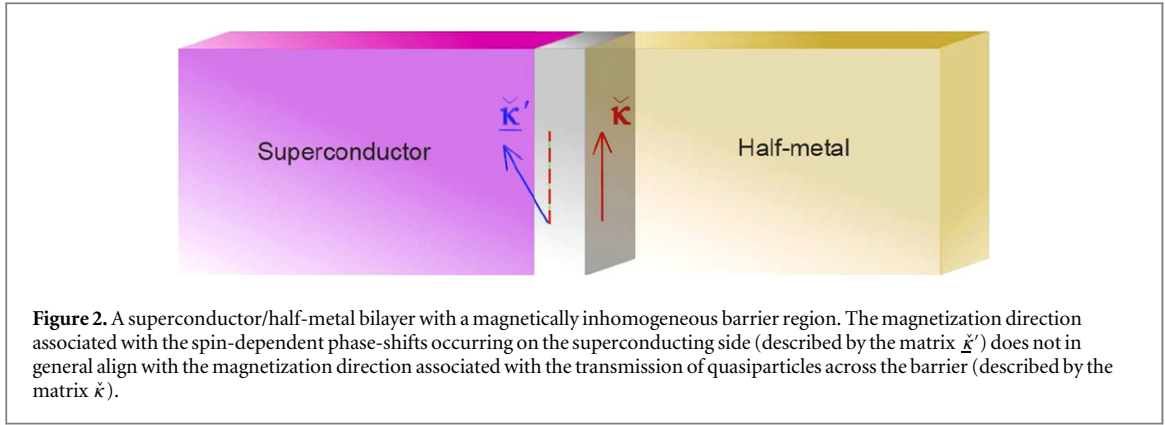
$$\mathcal{G}_{\chi}^{\mathrm{MR}} = \frac{1}{4}\mathcal{G}_q \sum_{nl} \vartheta_{nn}\mathcal{T}_{nl}\mathcal{P}_{nl}, \qquad \mathcal{G}_2^{\phi} = \frac{1}{2}\mathcal{G}_q \sum_{nn'} \vartheta_{nn'}^2 \tag{70d}$$

and $\mathcal{G}_{\underline{\chi}}^0, \mathcal{G}_{\underline{\chi}}^1, \mathcal{G}_{\underline{\chi}}^{\mathrm{MR}}$ are defined as $\mathcal{G}_{\chi}^0, \mathcal{G}_{\chi}^1$, and $\mathcal{G}_{\chi}^{\mathrm{MR}}$ with ϑ_{nn} replaced by $\underline{\vartheta}_{ll}$. Note that $\varphi_{nn'}$ and $\underline{\varphi}_{ll'}$ do not appear in these expressions, in accordance with the intuitive notion that scalar scattering phases should drop out in the quasiclassical limit, which operates with envelope functions only.

The case for only channel-conserving scattering (channel-diagonal problem) follows by taking in equations (69b)–(69d) and (70b)–(70d) only the terms with $n = l$. All other formulas (69a), (70a) remain unchanged. This case is treated in [55] to linear order in $\mathcal{P}_{nn}$, and our formulas reduce to these results for the considered limit. Note that for this case all spin-scalar phases cancel automatically and no averaging procedure over these phases is necessary.

5. Application for diffusive superconductor/half metal heterostructure

The problem of a superconductor in proximity contact with a half-metallic ferromagnet has been studied within the frameworks of Eilenberger equations [11, 12, 20, 50, 52, 67–69], Bogoliubov–de Gennes equations [70–73], recursive Green function methods [74], circuit theory [75], within a magnon-assisted tunneling model [76], and in the quantum limit [77]. Various experiments on superconductor/half-metal devices have been reported, both for layered systems involving high-temperature superconductors [78–81] and in diffusive structures involving conventional superconductors [82–88]. An important consequence of the new boundary conditions in equation (69a) is that half-metals can now be incorporated in the Usadel equation, which is appropriate to describe the second class of experiments mentioned above, whereas there previously existed no suitable boundary conditions to do so. Consider first a superconductor/half-metal bilayer with the interface located at $x = 0$ (see figure 2).

Figure 2. A superconductor/half-metal bilayer with a magnetically inhomogeneous barrier region. The magnetization direction associated with the spin-dependent phase-shifts occurring on the superconducting side (described by the matrix $\check{\underline{\kappa}}'$) does not in general align with the magnetization direction associated with the transmission of quasiparticles across the barrier (described by the matrix $\check{\kappa}$).

The superconductor is assumed to have a thickness well exceeding the superconducting coherence length. Our expansion parameters are the spin-dependent reflection phase shifts at the superconducting side of the interface, $\underline{\vartheta}_{ll'}$, and the tunneling probabilies $\mathcal{T}_{nl}$. For calculating triplet components in the half-metal it is sufficient to expand the solution for the Green function in the superconductor up to linear order, and the solution for the Green function in the half-metal up to quadratic order. The zeroth order term in the superconductor is pure spin-singlet, and the first-order term pure spin-triplet. Thus, up to and including the first order we can assume a bulk singlet order parameter, not affected by the interface scattering (corrections to the singlet order parameter arise only in second order in $\underline{\vartheta}_{ll'}$ and $\mathcal{T}_{nl}$). For future reference, we define the quantities $c \equiv \cosh(\nu) = -\mathrm{i}\frac{E}{\Omega}$, $s \equiv \sinh(\nu) = \mathrm{i}\frac{|\Delta|}{\Omega}$ with $\nu = \mathrm{atanh}(|\Delta|/E)$, $\Omega = \sqrt{|\Delta|^2 - E^2}$, and denote the SC phase as θ. We find for the triplet component $\underline{F}_{t0}$ in the superconductor

$$\underline{F}_{t0}(x) = \mathrm{i}\frac{\mathcal{G}^{\phi}cs}{\sigma_{\mathrm{SC}}\mathcal{A}q}\mathrm{e}^{\mathrm{i}\theta}\mathrm{e}^{-q|x|}\Big(\vec{\underline{m}}' \cdot \vec{\sigma}\Big)\mathrm{i}\sigma_y \tag{71}$$

with the normal-state conductivity $\sigma_{\mathrm{SC}} = 2e^2 N_{\mathrm{SC}} D_{\mathrm{SC}}$ in the superconductor (N_{SC} and D_{SC} are the normal-state density of states per spin projection at the Fermi level and the diffusion constant, respectively), contact area $\mathcal{A}$, and $q = \sqrt{2\Omega/\hbar D_{\mathrm{SC}}}$.

In the half-metal (width d), only spin-↑ particles have a non-zero density of states at the Fermi level. In the spirit of quasiclassical theory of superconductivity, a strong exchange field is incorporated not in the transport equation, but directly in the band structure which is integrated out at the quasiclassical level [17, 69], leaving only parameters such as the diffusion constant and normal state density of states at the Fermi level for each itinerant spin band. For transport in a half-metallic ferromagnet, this means one must just include one spin-band with diffusion constant D_{HM} in the Usadel equation. Thus, only the elements $G_{\uparrow\uparrow}$ and $F_{\uparrow\uparrow}$ exist in the Green function $\check{\mathbf{G}}$ of the half-metal. As we expand in the tunneling probability, we can (for energies well exceeding the Thouless energy $\hbar D_{\mathrm{HM}}/d^2$ of the half-metal) use the linearized Usadel equation,

$$\hbar D_{\mathrm{HM}}\partial_x^2 F_{\uparrow\uparrow} + 2\mathrm{i}E F_{\uparrow\uparrow} = 0. \tag{72}$$

Since there is only one anomalous Green function in the half-metal, we omit the spin indices for brevity of notation and define $F \equiv F_{\uparrow\uparrow}$. The general solution is $F(x) = A\mathrm{e}^{\mathrm{i}kx} + B\mathrm{e}^{-\mathrm{i}kx}$ with A, B being complex coefficients to be determined from the boundary conditions, and $k = \sqrt{2\mathrm{i}E/\hbar D_{\mathrm{HM}}}$. At the vacuum edge of the half-metal ($x = d$), we have $\partial_x F = 0$. At the interface between the superconductor and half-metal, the boundary conditions for F from the half-metallic side is obtained from equations (69a)–(70d) with $\mathcal{P}_{nl} = 1$. Note that for $\mathcal{P}_{nl} = 1$, we have $\mathcal{G}_{\underline{\chi}}^0 = \mathcal{G}_{\underline{\chi}}^1 = \mathcal{G}_{\underline{\chi}}^{\mathrm{MR}} \equiv \mathcal{G}_{\underline{\chi}}$ as well as $\mathcal{G}^0 = \mathcal{G}^1 = \mathcal{G}^{\mathrm{MR}}$. We find that in order to obtain a non-vanishing proximity effect, it is necessary that the magnetization direction associated with transmission across the barrier ($\check{\kappa}$) and spin-dependent phase-shifts picked up on the superconducting side of the interface ($\check{\underline{\kappa}}'$) are different. We set $\check{\kappa} = \check{\kappa}_z$ since the barrier magnetization determining the transmission properties is expected to be dominated by the half-metal magnetization which points in the z-direction. The boundary condition for F at $x = 0$ reads:

$$\sigma_{\mathrm{HM}}\mathcal{A}\partial_x F = 2\mathrm{i}\,\mathcal{G}_{\underline{\vartheta}}\,cs\mathrm{e}^{\mathrm{i}\theta}\Big(\underline{m}_x' - \mathrm{i}\underline{m}_y'\Big), \quad \mathcal{G}_{\underline{\vartheta}} = 2\mathcal{G}_{\underline{\chi}} + \frac{\mathcal{G}^{\phi}\mathcal{G}^0}{\sigma_{\mathrm{SC}}\mathcal{A}q} \tag{73}$$

with the normal-state conductivity $\sigma_{\mathrm{HM}} = e^2 N_{\mathrm{HM}} D_{\mathrm{HM}}$ in the half-metal (N_{HM} is the normal-state density of states at the Fermi level), and the conductance $\mathcal{G}_{\underline{\vartheta}}$ contains two terms: $2\mathcal{G}_{\underline{\chi}}$ which is proportional to $\sum_{nl}\underline{\vartheta}_{ll}\mathcal{T}_{nl}$, and a second term containing $\underline{\mathcal{G}}^{\phi}\mathcal{G}^0$ which is proportional to $(\sum_l \underline{\vartheta}_{ll})(\sum_{nl'}\mathcal{T}_{nl'})$. Moreover, $\underline{m}_x'$ and $\underline{m}_y'$ are the

normalized components of a possible misaligned barrier moment compared to the magnetization of the half-metal. We have taken this into account by writing:

$$
\hat{\underline{\kappa}}' = \underline{m}_x' \begin{pmatrix} \sigma_x & 0 \\ 0 & \sigma_x \end{pmatrix}_{\text{ph}} + \underline{m}_y' \begin{pmatrix} \sigma_y & 0 \\ 0 & \sigma_y^* \end{pmatrix}_{\text{ph}} + \underline{m}_z' \begin{pmatrix} \sigma_z & 0 \\ 0 & \sigma_z \end{pmatrix}_{\text{ph}} \tag{74}
$$

Inserting the general solution of *F* into the boundary conditions, one arrives at the final result for the proximity-induced superconducting correlations *F* in the half-metal:

$$
F(x) = -\frac{2\cosh[\mathrm{i}k(x-d)]}{\sinh(\mathrm{i}kd)} \frac{\mathcal{G}_{\underline{\vartheta}}\, cs}{\sigma_{\text{HM}}\mathcal{A}k} \mathrm{e}^{\mathrm{i}\theta}\big(\underline{m}_x' - \mathrm{i}\underline{m}_y' \big). \tag{75}
$$

This is the first time the Usadel equation has been used to describe the proximity effect in a superconductor/half-metal structure. Several observations can be made from the above expression. For small *E* the energy factors $c \propto E$ in the numerator and $k^2 \propto E$ in the denominator cancel, such that the proximity-effect, if present, happens even at $E = 0$. The proximity-effect is seen to be non-zero only if spin-dependent scattering phases at the superconducting side of the interface are present, and at the same time their quantization axis $\underline{\kappa}'$ is misaligned with that of the transmission amplitudes, κ. The reason for this is that phase-shifts on the half-metallic side are irrelevant on the quasiclassical level, because they are spin-scalar (only spin-↑ particles have a finite density of states there). On the other hand, the phase-shifts $\underline{\vartheta}_{nn}$ on the superconducting side have two consequences: they are responsible for an $\vec{S} \cdot \vec{m} = 0$ spin-triplet component on that side of the interface (where $\vec{S}$ is the spin vector of the Cooper pair), and they affect also transmission amplitudes. As a consequence, during transmission the quantization axis $\underline{\kappa}'$ can be rotated into the $S_z = \pm 1$ spin triplet components which are allowed to exist in the half-metal if spin-flip processes exist at the interface (e.g. due to some misaligned interface moments). This is exactly the reason why *F* also depends on $\underline{m}_x'$ and $\underline{m}_y'$ whereas it is independent on the barrier moment $\underline{m}_z'$: only a barrier moment with a component perpendicular to the magnetization of the half-metal can create spin-flip processes which rotate the $\vec{S} \cdot \vec{m} = 0$ into the $S_z = \pm 1$ components, and thus *F* also vanishes if $\underline{m}_x' = \underline{m}_y' = 0$.

Another important observation that can be made from the above expression is that a misaligned barrier moment effectively renormalizes the superconducting phase. Using spherical coordinates, we may write $\underline{m}_x' - \mathrm{i}\underline{m}_y' = \sin \underline{\Theta}' \mathrm{e}^{-\mathrm{i}\underline{\varphi}'}$ where $\underline{\varphi}'$ is the azimuthal angle describing the orientation of the barrier moment in the *xy*-plane. Thus, the effective phase becomes $\theta \to \theta - \underline{\varphi}'$. To see what consequence this has in terms of measurable quantities, we proceed to consider a Josephson junction with a half-metal by replacing the vacuum boundary condition at $x = d$ with another superconductor. Solving for the anomalous Green function *F* in the same way as above, we may compute the supercurrent flowing through the system via the formula (see equation (15)):

$$
I = \frac{eN_{\text{HM}}D_{\text{HM}}\mathcal{A}}{8} \int_{-\infty}^{\infty} \mathrm{d}E\, \mathrm{Tr}\Big\{ \hat{\tau}_3 \big(\check{\mathbf{G}}_{\text{HM}} \partial_x \check{\mathbf{G}}_{\text{HM}} \big)^K \Big\}. \tag{76}
$$

Here, Tr denotes a trace over 2×2 Nambu–Gor'kov space. After some calculations, one arrives at the result:

$$
I = I_0 \sin \underline{\Theta}_L' \sin \underline{\Theta}_R' \sin\Big(\theta_R - \theta_L + \underline{\varphi}_L' - \underline{\varphi}_R' \Big), \tag{77}
$$

where I_0 is a lengthy expression depending on parameters such as the width *d* of the half-metal and the temperature *T* (and which vanishes unless $\mathcal{G}_{\underline{\vartheta}}^L$ and $\mathcal{G}_{\underline{\vartheta}}^R$ are non-zero). To be general, we have allowed the spin-dependent phase-shifts for each superconductor and the barrier moment at each interface to be different, indicated by the notation '*L*' and '*R*' for left and right. We find that I_0 is negative, giving rise to a π-Josephson junction behavior for the case of $\underline{\varphi}_L' = \underline{\varphi}_R'$. Equation (77) is consistent with the ballistic case result of [12, 52, 89] and shows how a finite supercurrent will appear in a ring geometry even in the absence of any superconducting phase difference, $\theta_R - \theta_L = 0$, if the barrier moments are misaligned in the plane perpendicular to the junction, $\underline{\varphi}_L' - \underline{\varphi}_R' \neq 0$. A similar effect was also reported via circuit theory for a diffusive system [75], however not due to spin-dependent scattering phase shifts but due to some 'leakage terms'. Within our formalism, we thus obtain a so-called ϕ_0 Josephson junction behavior [90–94] with $\phi_0 = (\pi + \underline{\varphi}_L' - \underline{\varphi}_R') \,\mathrm{mod}\,(2\pi)$.

The above framework can be readily generalized to cover strongly spin-polarized ferromagnets building on the same idea as [17]. For a sufficiently large spin-splitting, the ↑- and ↓-conduction bands can be treated separately in the bulk with a separate Usadel equation for $F_{\uparrow\uparrow}$ and $F_{\downarrow\downarrow}$. These would then only couple via interface scattering and the strong exchange field would only enter by having different normal-state density of states $N_\uparrow$, $N_\downarrow$ and diffusion coefficients $D_\uparrow$, $D_\downarrow$ of the spin-bands in each separate Usadel equation.

6. Conclusions

We have derived new sets of boundary conditions for Usadel theory of superconductivity, appropriate for spin-polarized interfaces. We present a general solution of the problem appropriate for arbitrary transmission, spin-polarization, and spin-dependent scattering phases. The explicit equations for the most general set of boundary conditions are given in equations (33)–(34*b*), (38)–(40*b*), and (43)–(46). With the solution of this long-standing problem we anticipate a multitude of practical implementations in future to tackle superconducting systems that involve strongly spin-polarized materials. We have applied the general set of equations to various special cases important for practical use. We derived boundary conditions for an interface between a superconductor and a ferromagnetic insulator valid for arbitrary spin dependent scattering phases, equation (49). This extends previous work of [55], which was restricted to small scattering phases. Using an exact series expansion of the general set of boundary conditions, equations (50)–(55), we have obtained a perturbation series for the boundary conditions appropriate for such an interface, which allows for channel off-diagonal scattering and channel-dependent spin quantization axes, equations (57*a*) and (57*b*). For the tunneling limit, we have presented a new set of boundary conditions appropriate for arbitrary spin polarization, non-trivial spin texture across the interface, and allowing for channel off-diagonal scattering, equations (64*a*) and (64*b*). Neither of these three ranges of validity has been covered previously. As an application we then proceed to give a theoretical foundation of the boundary conditions used in [57–59], equations (69*a*)–(69*d*), which we have generalized for channel off-diagonal scattering and non-trivial spin texture across the interface. One central result of the application of our formalism is the extension of these relations to second order, including the important mixing terms between transmission and spin-dependent scattering phases. These terms, equations (70*a*)–(70*d*) generalize the corresponding terms from [55] to arbitrary spin polarization, possible nontrivial spin-texture across the interface, and channel off-diagonal scattering. We have demonstrated the application of the new set of boundary conditions by treating a diffusive superconductor/half-metal proximity junction and a diffusive superconductor/half-metal/superconductor Josephson junction. In the latter case we found a realization of a ϕ_0-junction. We are confident that our boundary conditions will advance the field of superconducting spintronics considerably.

Acknowledgments

Appendix A. Singular value decomposition of scattering matrix

We perform a singular value decomposition of the reflection and transmission matrices (with dimensions $n \times n$ for $\hat{S}_{11}$, $m \times m$ for $\hat{S}_{22}$, $n \times m$ for $\hat{S}_{12}$, and $m \times n$ for $\hat{S}_{21}$)

$$
\hat{\mathbf{S}} = \begin{pmatrix} \hat{S}_{11} & \hat{S}_{12} \\ \hat{S}_{21} & -\hat{S}_{22} \end{pmatrix}_{\times} = \begin{pmatrix} URV^{\dagger} & WT\check{Z}^{\dagger} \\ \check{W}\check{T}Z^{\dagger} & -\check{U}\check{R}\check{V}^{\dagger} \end{pmatrix}_{\times}.
\tag{A.1}
$$

Here $U, V, W, Z, \check{U}, \check{V}, \check{W}, \check{Z}$ are unitary matrices, and the $R, T, \check{R}, \check{T}$ contain the real and non-negative singular values in the main diagonal and are zero otherwise, i.e. $T^{\dagger} = T^{T}$ and $\check{T}^{\dagger} = \check{T}^{T}$, $R^{\dagger} = R$ and $\check{R}^{\dagger} = \check{R}$. We assume that the singular values are sorted from smallest to largest in R and $\check{R}$, and from largest to smallest in T and $\check{T}$. We introduce the unitary matrices $\Phi = W^{\dagger}U$, $\Psi = Z^{\dagger}V$ $\check{\Phi} = \check{W}^{\dagger}\check{U}$, and $\check{\Psi} = \check{Z}^{\dagger}\check{V}$. In terms of those, unitarity of the matrix $\hat{\mathbf{S}}$ requires that (we denote for simplicity the unit matrices $1_{n \times n}$ and $1_{m \times m}$ with the same symbol 1; the dimension is clear from the context)

$$\left(1 - R^2\right) \;=\; \Phi^\dagger T T^\dagger \Phi \;=\; \Psi^\dagger \check{T}^\dagger \check{T} \Psi \tag{A.2}$$

$$\left(1 - \check{R}^2\right) \;=\; \check{\Phi}^\dagger \check{T} \check{T}^\dagger \check{\Phi} \;=\; \check{\Psi}^\dagger T^\dagger T \check{\Psi}. \tag{A.3}$$

We see that $1 - R^2$ and $1 - \check{R}^2$ contain the eigenvalues of the Hermitian matrices on the right-hand sides of the equations, which requires that these eigenvalues coincide with the values in the diagonal matrices $TT^\dagger$, $\check{T}^\dagger \check{T}$, $\check{T}\check{T}^\dagger$, and $T^\dagger T$, respectively. Thus, with the sorting arrangement done above, the relations $(1 - R^2) \;=\; TT^\dagger \;=\; \check{T}^\dagger \check{T}$ and $(1 - \check{R}^2) \;=\; \check{T}\check{T}^\dagger \;=\; T^\dagger T$ hold. Because all singular values of T are real, this means that $\check{T} \;=\; T^\dagger$, $R \;=\; \sqrt{1 - TT^\dagger} \;=\; \sqrt{1 - \check{T}^\dagger \check{T}}$, $\check{R} \;=\; \sqrt{1 - T^\dagger T} \;=\; \sqrt{1 - \check{T}\check{T}^\dagger}$, and $R\check{T}^\dagger \;=\; T\check{R}$, $RT \;=\; \check{T}^\dagger \check{R}$. Furthermore, the unitary matrices Φ and Ψ commute with R and the unitary matrices $\check{\Phi}$ and $\check{\Psi}$ commute with $\check{R}$. In particular, those matrices are block diagonal, with block sizes given by the degeneracy of the singular values in R and $\check{R}$, respectively. The remaining unitarity requirements, using the above findings, reduce to

$$\Phi \Psi^\dagger \left(T\check{R} \right) \;=\; \left(T\check{R} \right) \check{\Psi} \check{\Phi}^\dagger \tag{A.4}$$

$$\Psi \Phi^\dagger \left(RT \right) \;=\; \left(RT \right) \check{\Phi} \check{\Psi}^\dagger. \tag{A.5}$$

That means that for the blocks corresponding to non-zero reflection singular values the above two equations lead to the one condition $\Phi^\dagger T \check{\Psi} \;=\; \Psi^\dagger T \check{\Phi}$. If there are no zero-reflection singular values then, remembering that Φ commutes with R and $\check{\Psi}$ with $\check{R}$,

$$\hat{\mathbf{S}} \;=\; \begin{pmatrix} U\Phi^\dagger & 0 \\ 0 & \check{U}\check{\Psi}^\dagger \end{pmatrix}_{\times} \begin{pmatrix} R & T \\ T^\dagger & -\check{R} \end{pmatrix}_{\times} \begin{pmatrix} \Phi V^\dagger & 0 \\ 0 & \check{\Psi}\check{V}^\dagger \end{pmatrix}_{\times}. \tag{A.6}$$

The blocks with zero-reflection singular values can be treated separately, and it is easily seen that the singular value decomposition of the scattering matrix has the general form

$$\hat{\mathbf{S}} \;=\; \begin{pmatrix} \mathcal{U} & 0 \\ 0 & \check{\mathcal{U}} \end{pmatrix}_{\times} \begin{pmatrix} \sqrt{1 - TT^\dagger} & T \\ T^\dagger & -\sqrt{1 - T^\dagger T} \end{pmatrix}_{\times} \begin{pmatrix} \mathcal{V}^\dagger & 0 \\ 0 & \check{\mathcal{V}}^\dagger \end{pmatrix}_{\times} \tag{A.7}$$

with unitary matrices $\mathcal{U}$, $\check{\mathcal{U}}$, $\mathcal{V}$, and $\check{\mathcal{V}}$. The decomposition is not unique.

Appendix B. Polar decomposition of scattering matrix

An important feature of the above representation is that the center matrix is Hermitian. If we only require this property of the central part, but not necessarily diagonality of the $m \times n$ matrix T, then we can find an entire class of transformations that keep this property. We define $\mathcal{R} D \check{\mathcal{R}}^\dagger \;=\; T$ with unitary matrices $\mathcal{R}$ and $\check{\mathcal{R}}$. Then

$$\hat{\mathbf{S}} \;=\; \begin{pmatrix} \mathcal{U}\mathcal{R} & 0 \\ 0 & \check{\mathcal{U}}\check{\mathcal{R}} \end{pmatrix}_{\times} \begin{pmatrix} \sqrt{1 - DD^\dagger} & D \\ D^\dagger & -\sqrt{1 - D^\dagger D} \end{pmatrix}_{\times} \begin{pmatrix} \mathcal{R}^\dagger \mathcal{V}^\dagger & 0 \\ 0 & \check{\mathcal{R}}^\dagger \check{\mathcal{V}}^\dagger \end{pmatrix}_{\times} \tag{B.1}$$

where D is now an $n \times m$ matrix that is not necessarily diagonal anymore. Consider now some special cases. First, we chose $\mathcal{R} \;=\; \mathcal{V}^\dagger$, $\check{\mathcal{R}} \;=\; \check{\mathcal{V}}^\dagger$. Then

$$\hat{\mathbf{S}} \;=\; \begin{pmatrix} \mathcal{U}V^\dagger & 0 \\ 0 & \check{\mathcal{U}}\check{\mathcal{V}}^\dagger \end{pmatrix}_{\times} \begin{pmatrix} \sqrt{1 - C'C'^\dagger} & C' \\ C'^\dagger & -\sqrt{1 - C'^\dagger C'} \end{pmatrix}_{\times} \tag{B.2}$$

with $C' \;=\; \mathcal{V}T\check{\mathcal{V}}^\dagger$ gives a polar decomposition of the reflection parts of the scattering matrix $\hat{\mathbf{S}}$. Similarly, $\mathcal{R} \;=\; \mathcal{U}^\dagger$, $\check{\mathcal{R}} \;=\; \check{\mathcal{U}}^\dagger$ leads to

$$\hat{\mathbf{S}} \;=\; \begin{pmatrix} \sqrt{1 - CC^\dagger} & C \\ C^\dagger & -\sqrt{1 - C^\dagger C} \end{pmatrix}_{\times} \begin{pmatrix} \mathcal{U}V^\dagger & 0 \\ 0 & \check{\mathcal{U}}\check{\mathcal{V}}^\dagger \end{pmatrix}_{\times} \tag{B.3}$$

with $C \;=\; \mathcal{U}T\check{\mathcal{U}}^\dagger \;=\; \mathcal{U}V^\dagger C' (\check{\mathcal{U}}\check{\mathcal{V}}^\dagger)^\dagger$. We can also chose a decomposition in the form

$$\hat{\mathbf{S}} \;=\; \begin{pmatrix} \mathcal{U}\mathcal{V}^\dagger & 0 \\ 0 & 1 \end{pmatrix}_{\times} \begin{pmatrix} \sqrt{1 - C''C''^\dagger} & C'' \\ C''^\dagger & -\sqrt{1 - C''^\dagger C''} \end{pmatrix}_{\times} \begin{pmatrix} 1 & 0 \\ 0 & \check{\mathcal{U}}\check{\mathcal{V}}^\dagger \end{pmatrix}_{\times} \tag{B.4}$$

with $C'' \;=\; \mathcal{V}T\check{\mathcal{U}}^\dagger$, or other decompositions.

These decompositions are unique when there are no zero-reflection singular values. This means, that under the conditions of no zero-reflection channels $\mathcal{U}V^{\dagger}$ and $\check{\mathcal{U}}\check{\mathcal{V}}^{\dagger}$ are uniquely defined, as the matrices C and D are. The unique unitary matrices $\mathcal{U}V^{\dagger}$ and $\check{\mathcal{U}}\check{\mathcal{V}}^{\dagger}$ are the surface scattering matrices $\mathcal{S}$ and $\check{\mathcal{S}}$, that occur in the limit of zero transmission equation (19).

Appendix C. Parameterization of scattering matrix

We now turn to a useful parameterization of the transmission matrix C. We note that with the definition

$$C = \left(1 + tt^{\dagger}\right)^{-1} 2t \tag{C.1}$$

we obtain

$$\begin{pmatrix} \sqrt{1 - CC^{\dagger}} & C \\ C^{\dagger} & -\sqrt{1 - C^{\dagger}C} \end{pmatrix}_{\times} = \begin{pmatrix} \hat{r} & \hat{d} \\ \hat{d}^{\dagger} & -\check{\check{r}} \end{pmatrix}_{\times} \tag{C.2}$$

with

$$\hat{r} = \left(1 + tt^{\dagger}\right)^{-1}\left(1 - tt^{\dagger}\right) \tag{C.3}$$

$$\check{\check{r}} = \left(1 + t^{\dagger}t\right)^{-1}\left(1 - t^{\dagger}t\right) \tag{C.4}$$

$$\hat{d} = \left(1 + tt^{\dagger}\right)^{-1} 2t. \tag{C.5}$$

To connect with the main text, we have $t = \pi\hat{t}$. Furthermore, if $t = u\theta v^{\dagger}$ is a singular decomposition for t, then $C = u\left[(1 + \theta^2)^{-1}2\theta\right]v^{\dagger}$ is a singular decomposition of C. Conversely, if $C = u\delta v^{\dagger}$ is a singular decomposition for C, then $t = u\left[(1 - \sqrt{1 - \delta^2})\big/\delta\right]v^{\dagger}$ is a singular decomposition for t. If $0 < \theta < 1$ then $0 < \delta < 1$ and vice versa. Thus, the parameterization in terms of t is equivalent to that in terms of C.

Appendix D. Expansion to third order of equation 56

To third order we obtain from equation (56)

$$\check{\mathcal{I}}^{(1)} = \mathrm{i}\left[\check{K}, \check{G}\right] \tag{D.1}$$

$$\check{\mathcal{I}}^{(2)} = -\frac{1}{2}\left[\check{K}^2, \check{G}\right] + \mathrm{i}\left[\check{K}, \left(\check{G} + \mathrm{i}\pi\check{1}\right) \circ (\check{G}')^{(1)} \circ \check{G}\right] \tag{D.2}$$

$$\begin{aligned}
\check{\mathcal{I}}^{(3)} = {}&{-}\frac{\mathrm{i}}{6}\left[\check{K}^3, \check{G}\right] - \frac{1}{2}\left[\check{K}^2, \left(\check{G} + \mathrm{i}\pi\check{1}\right) \circ (\check{G}')^{(1)} \circ \check{G}\right] \\
&{+} \mathrm{i}\left[\check{K}, \left(\check{G} + \mathrm{i}\pi\check{1}\right) \circ (\check{G}')^{(2)} \circ \check{G}\right] \\
&{+} \mathrm{i}\left[\check{K}, \left(\check{G} + \mathrm{i}\pi\check{1}\right) \circ (\check{G}')^{(1)} \circ \check{G} \circ (\check{G}')^{(1)} \circ \check{G}\right]
\end{aligned} \tag{D.3}$$

and

$$(G')^{(1)} = -\frac{\mathrm{i}}{2\pi^2}\left[\check{K}, \check{G}\right], \quad (G')^{(2)} = -\frac{1}{4\pi^2}\left[\check{K}^2, \check{G}\right]. \tag{D.4}$$

This can be simplified further noting

$$\check{G} \circ (\check{G}')^{(1)} = -(\check{G}')^{(1)} \circ \check{G}, \tag{D.5}$$

$$2\pi^2(\check{G}')^{(1)} \circ (\check{G}')^{(1)} = -\left\{(\check{G}')^{(2)}, \check{G}\right\}, \tag{D.6}$$

$$\left(\check{G} + \mathrm{i}\pi\check{1}\right) \circ \check{G} = \mathrm{i}\pi\left(\check{G} + \mathrm{i}\pi\check{1}\right), \tag{D.7}$$

$$2\pi^2\left[\check{K}^n, (\check{G}')^{(1)}\right] = -\mathrm{i}\left[\check{K}^{n+1}, \check{G}\right], \tag{D.8}$$

$$4\pi^2\left[\check{K}, (\check{G}')^{(2)}\right] = -\left[\check{K}^3, \check{G}\right], \tag{D.9}$$

yielding equation (57a) of the main text.

Appendix E. Expansion of matrix current for finite transmission

From section 4.3 we obtain the following expressions to second order in the spin dependent reflection phases and in the transmission probability:

$$\check{\mathcal{I}}^{(1)} = -2\pi i \left[\check{t}^o, \check{g}_0^o \right]_\circ^{(1)} + i \left[\check{K}, \check{G} \right], \tag{E.1}$$

$$\check{\mathcal{I}}^{(2)} = -2\pi i \left[\check{t}^o, \check{g}_0^o \right]_\circ^{(2)} + i \left[\check{K}, \left(\check{g}^i \right)^{(1)} \right] - \frac{1}{2} \left[\check{K}^2, \check{G} \right], \tag{E.2}$$

with

$$\left[\check{t}^o, \check{g}_0^o \right]_\circ^{(1)} = \left[\check{\tau}_0 \underline{\check{G}} \check{\tau}_0^\dagger, \check{G} \right]_\circ \tag{E.3}$$

$$\left[\check{t}^o, \check{g}_0^o \right]_\circ^{(1)} = \left[\check{G}_1, \check{G} \right]_\circ \tag{E.4}$$

$$\left[\check{t}^o, \check{g}_0^o \right]_\circ^{(2)} = \left[\check{\tau}_0 \left(\underline{\check{g}}_0^i \right)^{(1)} \check{\tau}_0^\dagger, \check{G} \right]_\circ + \left[\check{G}_1, \left(\check{g}_0^o \right)^{(1)} \right]_\circ$$
$$+ \check{G}_1 \circ \check{G} \circ \left[\check{G}_1, \check{G} \right]_\circ + \left[\check{G}_1, \check{G} \right]_\circ \circ \check{G} \circ \check{G}_1$$
$$+ \frac{i}{2} \left(\left[\left[\check{K}, \check{G}_1 \right], \check{G} \right]_\circ + \left[\tau_0 \left[\underline{\check{K}}, \underline{\check{G}} \right] \tau_0^\dagger, \check{G} \right]_\circ \right), \tag{E.5}$$

and

$$\left(\check{g}^i \right)^{(1)} = \left(\check{G} + i\pi \check{1} \right) \circ \left(\left[\check{G}_1, \check{G} \right]_\circ + (G')^{(1)} \circ \check{G} \right) \tag{E.6}$$

$$\left(\underline{\check{g}}_0^i \right)^{(1)} = \left(\underline{\check{G}} + i\pi \underline{\check{1}} \right) \circ \left(\underline{\check{G}}' \right)^{(1)} \circ \underline{\check{G}} \tag{E.7}$$

$$\left(\check{g}_0^o \right)^{(1)} = \left(\check{G} - i\pi \check{1} \right) \circ \left(\check{G}'' \right)^{(1)} \circ \check{G} \tag{E.8}$$

with $(\check{G}'')^{(1)} = -(\check{G}')^{(1)}$ from equation (D.4). Collecting everything together, we obtain the result shown in equations (63*a*) and (63*b*) of the main text.

Appendix F. Term of second order in transmission probability

For completeness we present here the expression of order $\mathcal{T}_{nl}^2$:

$$I_4 = \mathcal{G}_4^0 \, \underline{\check{G}} \circ \check{G} \circ \underline{\check{G}} + \mathcal{G}_4^1 \, \check{\kappa} \underline{\check{G}} \check{\kappa} \circ \check{G} \circ \check{\kappa} \underline{\check{G}} \check{\kappa}$$
$$+ \mathcal{G}_4^{\mathrm{MR}} \left(\underline{\check{G}} \circ \check{G} \circ \check{\kappa} \underline{\check{G}} \check{\kappa} + \check{\kappa} \underline{\check{G}} \check{\kappa} \circ \check{G} \circ \underline{\check{G}} \right) + \mathcal{G}_4^{\mathrm{MR}'} \left\{ \check{\kappa}, \underline{\check{G}} \right\} \circ \check{G} \circ \left\{ \check{\kappa}, \underline{\check{G}} \right\}$$
$$+ \mathcal{G}_4^{\mathrm{mix}} \left(\underline{\check{G}} \circ \check{G} \circ \left\{ \check{\kappa}, \underline{\check{G}} \right\} + \left\{ \check{\kappa}, \underline{\check{G}} \right\} \circ \check{G} \circ \underline{\check{G}} \right)$$
$$+ \mathcal{G}_4^{\mathrm{mix}'} \left(\check{\kappa} \underline{\check{G}} \check{\kappa} \circ \check{G} \circ \left\{ \check{\kappa}, \underline{\check{G}} \right\} + \left\{ \check{\kappa}, \underline{\check{G}} \right\} \circ \check{G} \circ \check{\kappa} \underline{\check{G}} \check{\kappa} \right) \tag{F.1}$$

with

$$\mathcal{G}_4^0 = \frac{1}{8} \mathcal{G}_q \sum_{nln'l'} p_{nln'l'} \mathcal{T}_{nl} \mathcal{T}_{n'l'} \left(1 + \sqrt{1 - \mathcal{P}_{nl}^2} \right) \left(1 + \sqrt{1 - \mathcal{P}_{n'l'}^2} \right) \tag{F.2}$$

$$\mathcal{G}_4^1 = \frac{1}{8} \mathcal{G}_q \sum_{nln'l'} p_{nln'l'} \mathcal{T}_{nl} \mathcal{T}_{n'l'} \left(1 - \sqrt{1 - \mathcal{P}_{nl}^2} \right) \left(1 - \sqrt{1 - \mathcal{P}_{n'l'}^2} \right) \tag{F.3}$$

$$\mathcal{G}_4^{\mathrm{MR}} = \frac{1}{8} \mathcal{G}_q \sum_{nln'l'} p_{nln'l'} \mathcal{T}_{nl} \mathcal{T}_{n'l'} \left(1 + \sqrt{1 - \mathcal{P}_{nl}^2} \right) \left(1 - \sqrt{1 - \mathcal{P}_{n'l'}^2} \right) \tag{F.4}$$

$$\mathcal{G}_4^{\mathrm{MR}'} = \frac{1}{8} \mathcal{G}_q \sum_{nln'l'} p_{nln'l'} \mathcal{T}_{nl} \mathcal{T}_{n'l'} \mathcal{P}_{nl} \mathcal{P}_{n'l'} \tag{F.5}$$

$$\mathcal{G}_4^{\mathrm{mix}} = \frac{1}{8} \mathcal{G}_q \sum_{nln'l'} p_{nln'l'} \mathcal{T}_{nl} \mathcal{T}_{n'l'} \mathcal{P}_{n'l'} \left(1 + \sqrt{1 - \mathcal{P}_{nl}^2} \right) \tag{F.6}$$

$$\mathcal{G}_4^{\mathrm{mix}'} = \frac{1}{8} \mathcal{G}_q \sum_{nln'l'} p_{nln'l'} \mathcal{T}_{nl} \mathcal{T}_{n'l'} \mathcal{P}_{n'l'} \left(1 - \sqrt{1 - \mathcal{P}_{nl}^2} \right) \tag{F.7}$$

with $p_{nln'l'} \equiv \delta_{nn'} + \delta_{ll'} - \delta_{nn'}\delta_{ll'}$, arising from averaging over the typical phase factor $\mathrm{e}^{\mathrm{i}(\psi_{nl} - \psi_{n'l} + \psi_{n'l'} - \psi_{nl'})}$ of spin-scalar transmission phases. The channel-diagonal case follows from setting $n = l = n' = l'$ and $p_{nnnn} = 1$.

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