nature reviews physics

https://doi.org/10.1038/s42254-023-00632-w

Review article

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# The superconducting diode effect

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# Supplementary Information: The superconducting diode effect

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## Nonreciprocity, symmetry constraints, and magnetochirality

In condensed matters, nonreciprocity refers to the spatial dependence of physical quantities. A prototypical example of nonreciprocal transport is a diode effect which refers to highly direction-selective electron transport in systems with a lack of spatial inversion center. Until recently, nonreciprocity was thought to be a transport phenomenon associated with dissipative materials. For instance, in conventional semiconductors, where resistance is the nonreciprocal quantity, nonreciprocity refers to charge transport that is sensitive to the polarity of current or bias potential. Such nonreciprocal charge transport leads to a diode effect in a spatially asymmetric pn junction[1,2], in which, spatial asymmetry of the junction is associated with electron-hole asymmetry across the contact of n- and p-type semiconductors.

In modern quantum condensed matter physics, in addition to electron-hole asymmetric junctions, nonreciprocal charge transport can also be induced in spatially symmetric devices, in which transport is direction-selective when space-inversion and/or time-reversal symmetry are broken. Space-inversion symmetry is either intrinsically broken or can be broken by applying an electric field externally. Similarly, time-reversal symmetry can be broken either by applying an external magnetic field or through intrinsic magnetization, leading to an observation of field-free SDE[3–9]. Here, it is highlighted how the nonreciprocity of supercurrent associates with the nonreciprocal behavior of physical quantities characterizing current-voltage (I-V) and current-phase relation (CPR), e.g., resistance and inductance respectively.

### Nonreciprocity of supercurrent

In 1996, before even prediction/observation of nonreciprocity in normal (semi)conductors by Rikken et al.[10,11], V. M. Edelstein[12] proposed nonreciprocity in the critical supercurrent. Followed by his earlier work characterizing Cooper pairing in noncentrosymmetric superconductors[13] and describing magnetoelectric effects in polar superconductors[14], V. M. Edelstein[12] proposed that if the mixed product $(\boldsymbol{r}\times\boldsymbol{B})\cdot\hat{\boldsymbol{j}}$ is non-vanishing in polar superconductors, then the magnitude of the critical current $j_c(B)$ depends on the sign of this mixed product, i.e., the critical current appears to be different for two opposite directions. That is, the nonreciprocity of supercurrent can be characterized by the magnetic field-dependent critical current as

$$j_c(B) = j_c(0)[1+\gamma_j(\boldsymbol{r}\times\boldsymbol{B})\cdot\hat{\boldsymbol{j}}] \tag{1}$$

Here $r$ is the unit vector along the polar axis, $\hat{\boldsymbol{j}}$ is the unit vector along the supercurrent, $\gamma_j$ is the coefficient of magnetochiral anisotropy (MCA), and $B$ is an in-plane magnetic field. The exact expression for the observable (coefficient of supercurrent MCA) $\gamma_j$, derived by employing GL theory for a film of polar superconductor and showing its dependence on the coherence length, Fermi energy, SOI energy, and the upper critical field, etc., can be found in the reference[12].

#### *From resistance to supercurrent*

Rikken et al.[10,11] generalized Onsager's reciprocal theorem to the nonlinear regime and gave a heuristic argument for nonreciprocity and MCA in two-dimensional chiral[10] and polar diffusive[11] conductors. In their seminal proposal of MCA in polar diffusive (semi)conductors, Rikken et al.[11] suggested that nonreciprocal nonlinear resistive response, characterized by the directional I-V characteristics, can be described by a current-dependent resistance $R(I)$ as

$$R(I) = R_0[1+\beta B^2+\gamma_R(\boldsymbol{B}\times\boldsymbol{r})\cdot\boldsymbol{I}] \tag{2}$$

Here R, B, and I are the resistance, magnetic field, and electric current, respectively. The unit vector $\boldsymbol{r}$ represents the direction along which mirror symmetry is broken. On the right-hand side, the first term is the resistance at zero magnetic fields, the second term denotes the normal magnetoresistance, and the third term corresponds to the MCA. Such nonlinear nonreciprocal resistive transport, caused by current-dependent resistance or nonlinear voltage-drop, can be detected by measuring the second harmonic signal through lock-in techniques, further details are presented later in the Review.

Current-dependent resistance (Eq. 2) and the magnetic-field dependent critical current (Eq. 1) demonstrate that non-reciprocity and MCA can be observed in (semi)conductors[10,11,15–19] that allow resistive current transport as well as in superconductors[12,14,20–23] that display dissipationless supercurrent transport. So a question arises naturally: how nonreciprocity can uniquely be defined in these two systems with completely contrasting transport behaviour? As pointed out by Rikken et al.[10,11], when both inversion and time-reversal symmetries are broken, the finite MCA coefficient $\gamma$ gives rise to different resistance for electric currents traversing in different (opposite) directions. That is, MCA can be defined as the inequivalence of $R(+I)$ and $R(-I)$. In (semi)conductors, resistances along opposite directions differ, i.e. $R(+I) \neq R(-I)$, but both $R(+I)$ and $R(-I)$ normally take finite values. On the other hand, in superconductors, such a situation becomes more drastic: either one of $R(\pm I)$ remains finite while the other completely vanishes. With this consideration in superconductors, it becomes more appropriate to define nonreciprocity in terms of (super)current. That is, nonreciprocity in superconductors means supercurrent flows along one direction while normal current along the other(opposite). Observation of such a situation is more probable near critical temperature $T_c$, i,e., in the fluctuation regime of metal-superconductor resistive transition, where the critical current is different along opposite directions, i.e. $I_{c+} \neq I_{c-}$. Thus, if the current is tuned between $I_{c+}$ and $I_{c-}$, the system displays zero resistance for the supercurrent but nonzero for the normal current.

It can be understood how conductance varies while going from normal to a superconducting phase. The linear resistance $R_0$ is normally scaled by the Fermi energy $E_F$, i.e., the kinetic energy of the electrons, while the MCA coefficient $\gamma$ depends upon the strength of SOI and the magnetic field. Correspondingly, nonlinear resistance induced by MCA may be treated as a perturbation to $R_0$. In the normal conducting phase, because the SOI energy ($E_{soi}$) and the Zeeman energy ($\mu_B B$) is usually much smaller (by many orders of magnitude) than $E_F$, MCA coefficient $\gamma \rightarrow \gamma_N$ is typically very tiny, usually of the order of $\sim 10^{-3}$ to $10^{-2}$ T$^{-1}$ A$^{-1}$ in typical metals[10,16,20]. However, as the superconducting phase develops, superconducting transition temperature $T_c$ or the superconducting gap $\Delta_{sc}$ appears as a new energy scale. That is, the energy scale in superconductors, to which the strength of SOI has to be compared with, is a superconducting gap and not the Fermi energy. Since the energy scale ($\sim$meV) in the superconductors is much smaller than the Fermi energy ($\sim$eV) in metals, the effects of SOI and Zeeman energy greatly enhance in the superconducting phase[20,24]. As a result, near the superconducting transition temperature $T \gtrsim T_c$, the MCA coefficient becomes reasonably large[25] and, thus, the paraconductivity[26] above $T_c$ becomes nonreciprocal. In the superconducting fluctuation region, i.e. when $T \rightarrow T_c$ and the superconducting order parameter $\Delta_{sc}$ develops, a sizable enhancement in MCA coefficient $\gamma_S$ is found (ref.[20,22,24,25]) and robust non-reciprocal charge transport is demonstrated in noncentrosymmetric superconductors[20,23]. For instance, by employing GL theory for an Ising type superconductor $MoS_2$, R. Wakatsuki et al.[20] showed that the ratio of MCA coefficients in the superconducting resistive region ($\gamma_S$) and the normal resistive region ($\gamma_N$) is quite large

$$\frac{\gamma_S}{\gamma_N} \sim \left(\frac{E_F}{k_B T_c}\right)^3 \tag{3}$$

Such anomalous enhancement of the MCA coefficient, as it is associated with the energy scale difference between the superconducting gap and the Fermi energy, can be considered an intrinsic feature of both Rashba and Ising type noncentrosymmetric superconductors[24]. However, mainly due to a gradual decrease in the linear resistance $R_0$ during the metal-superconducting transition, $R_0$ remains larger (by orders of magnitude) than the nonlinear resistance in low-dimensional superconducting materials such as $MoS_2$ (ref.[20]), $WS_2$ (ref.[21]) and $Bi_2Te_3$/FeTe (ref.[22]). As a result, the low rectification ratio in these superconducting materials does not suffice for device implementation. In this regard, it motivated several research groups to search for exotic materials and novel mechanisms/principles to enlarge the rectification effect and guide the design of efficient SDE.

### ***From inductance to supercurrent***

Nonreciprocity in the fluctuation regime of metal-superconductor resistive transition confines SDE to a narrow temperature window near $T_c$. Baumgartner et al.[27] pointed out that the temperature window in which MCA coefficient becomes sizeable must be widened for a sustainable fabrication of devices showing SDE. To achieve this milestone, the authors demonstrated supercurrent rectification in the superconducting phase, i.e., far below the transition temperature $T_c$. Since d.c. measurement of resistance–current (R–I) curve is not viable at low temperatures, as the resistance vanishes, supercurrent response to an alternating-current (a.c.) excitation is studied, which is described by its superfluid stiffness, and thus, can be detected through kinetic inductance measurements.

If mirror symmetry is broken along an out-of-plane direction ($\hat{e}_z$), whereas the current I and magnetic field B are directed in-plane, MCA or nonreciprocity for the superfluid can be described by an equation similar to that for the current-dependent resistance in polar superconductors (2), i.e.,

$$L(I) = L_0[1 + \gamma_L \hat{e}_z(\boldsymbol{B} \times \boldsymbol{I})] \tag{4}$$

Here resistance ($R$) is substituted for the kinetic inductance ($L$). The nonreciprocity in supercurrent could then be characterized by a new observable, i.e., MCA coefficient $\gamma_L$, and can be quantified by measuring asymmetry in CPR. Further details on this mechanism are presented later in the Review.

#### *Nonreciprocity in chiral conductors*

In chiral conductors, finite MCA leads to a nonreciprocal transport when a component of the applied magnetic field is parallel to the current, i.e., handed-selective resistance $R^{D/L}$ depends on the relative orientation of magnetic field and current as[10]:

$$R^{D/L}(I) = R_0[1 + \beta B^2 + \gamma^{D/L} \boldsymbol{B} \cdot \boldsymbol{I}] \tag{5}$$

where $D/L$ denotes the right/left-handedness of chiral conductors and the parity reversal symmetry requires that $\gamma^D = -\gamma^L$, i.e, such an MCA coefficient remains finite only for chiral conductors. The handed-selective nonreciprocal transport has been observed in the normal chiral conductors, e.g., Bi helices[10], carbon nanotubes[15,28], bulk organic conductors[16], chiral magnets[29,30], and elemental trigonal tellurium (t-Te)[31,32]. Furthermore, finite handed-selective MCA has been observed in chiral superconductors, e.g., Ru-$Sr_2RuO_4$ eutectic system[33,34] and $WS_2$ nanotubes[21].

## Measuements of superconducting diode effect

Based on the working temperature, or a working regime of phase diagram representing metal-superconductor resistive transition, observation of SDE can be classified into two main categories: (i) SDE based on the nonreciprocity of depairing critical current near the superconducting transition temperature ($T \approx T_c$), i.e., in the fluctuation regime of metal-superconductor resistive transition, and (ii) SDE based on the nonreciprocity of supercurrent at sub-Kelvin temperatures ($T \ll T_c$), i.e., deep in the superconducting phase regime.

### Magnetochiral anisotropy of the resistance

In the fluctuation regime of resistive transition close to $T_c$, SDE can be described by MCA of the resistance ($\gamma_R \to \gamma_S$, as defined in equation (2)), similar to that in semiconductors, and may be characterized by I-V curves. In this regime, the MCA coefficient $\gamma_S$ can be found by measuring the second harmonic signal in lock-in measurements. That is, for an ac current ($I_{in} = I \sin \omega t$) with an amplitude of $I$ and a frequency of $\omega$ applied as input, the nonlinear voltage-drop and current-dependent resistance can be derived from the nonlinear resistance term in equation (2) as:

$$\begin{aligned} V_{2\omega}(t) &= \gamma_S B R_\omega I^2 \sin^2 \omega t \\ &= \frac{1}{2}\gamma_S B R_\omega I^2 \left[1 + \sin\left(2\omega t - \frac{\pi}{2}\right)\right] \\ R_{2\omega} &= \frac{1}{2}\gamma_S B R_\omega I \end{aligned} \tag{6}$$

Here $R_\omega$ corresponds to the current-independent linear resistance $R_0$, while $R_{2\omega}$ represents the second-order nonlinear resistance, which is dependent on both the current and the magnetic field. Thus by measuring the first- ($R_\omega$) and second-harmonic $R_{2\omega}$ sheet/junction resistances through $2\omega$ voltage response, $\gamma_S$ can be estimated as $\gamma_S = \frac{2R_{2\omega}}{BIR\omega}$.

However, such resistive measurements cannot realistically simulate the intrinsic SDE at temperatures well below $T_c$ due to no measurable resistance in this regime ($R_0 = 0$). Thus, the efficiency of SDE is expected to be finite only at $T \approx T_c$ while negligibly small both at temperatures well below $T_c$ and above $T_c$ ($\gamma_N \ll \gamma_S$). For instance, as shown in Ref.[35], MCA coefficient $\gamma_S$ shows a sharp increase in the fluctuation regime and reaches its maximal value $\gamma_S \simeq 550$ T$^-$A$^-$ at $T_c$, however, $\gamma_S$ remains negligibly small at temperatures well below $T_c$. Though the observation seems to be at variance with the theoretical predictions for intrinsic SDE[36–39] and the temperature dependence of experimentally measured MCA in JJs[27,40], it is an expected outcome of resistive measurements. On the other hand, as shown for JJs[27], finite MCA coefficient $\gamma_S \simeq 4.1 \times 10^6$ T$^-$A$^-$ observed through resistive measurements near $T_c \sim 1.45$ K is of the same order (namely, in the range of $10^6$ T$^-$A$^-$) of the corresponding MCA coefficient observed for the inductance (measured at $T = 100$ mK), $\gamma_L \simeq 0.77 \times 10^6$ T$^-$A$^-$.

### Magnetochiral anisotropy of the inductance

Unlike the fluctuation regime, where the nonreciprocity of depairing critical current is tied to the nonlinear resistance, the nonreciprocity of sub-Kelvin supercurrent promises a fully superconducting/dissipationless nonreciprocal circuit element. Deep in the sub-Kelvin superconducting regime of the phase diagram, i.e., far below the transition temperature where resistance is zero (so DC measurements are not feasible), supercurrent MCA and a corresponding SDE (supercurrent rectification/nonreciprocity) are characterized rather by measuring kinetic (or Josephson) inductance (clearly with AC measurements). By measuring Josephson inductance, nonreciprocal supercurrent can be linked to an asymmetry in the CPR, induced by the simultaneous breaking of inversion and time-reversal symmetry such that B is not parallel to I, and the MCA coefficient ($\gamma_L$) for the supercurrent can be directly derived from the equation (4).

This mechanism can be understood from a semiquantitative model[27,41,42] in which Josephson inductance can be derived from the CPR relation $I = I_{c0} f(\varphi)$ (where f is a $2\varphi$-periodic function) and second Josephson equation $\dot{\varphi} = 2\pi V/\Phi_0$ (where $\Phi_0 = h/(2e)$ is the magnetic flux quantum) as

$$L(I) = \frac{V}{\frac{dI}{dt}} = \frac{V}{\frac{dI}{d\varphi}\dot{\varphi}} = \frac{\Phi_0}{2\pi I_{c0}\frac{df(\varphi)}{d\varphi}} = \frac{\Phi_0}{2\pi}\left[\frac{dI(\varphi)}{d\varphi}\right]^{-1} \tag{7}$$

It shows that Josephson inductance is a convenient probe to study CPR symmetry by investigating the effects of space-inversion/time-reversal symmetry breaking on the CPR. Let's assume a JJ configuration in which electric current is flowing along the x-direction, while inversion and time-reversal symmetry is broken by applying out-of-plane electric field $\mathbf{E} = E_z\hat{z}$ and in-plane magnetic field $\boldsymbol{B}_{ip} = B_x\hat{x} + B_y\hat{y}$, respectively.

Equation (7) shows that $L(I)$ is inversely proportional to the derivative of the CPR, therefore, the minimum of Josephson inductance occurs at the inflection-point of the CPR. In the absence of an in-plane magnetic field component along the y-direction ($B_y = 0$), CPR remains symmetric around inflection-point appearing at zero-phase, that is $(i,\varphi) = (0,0)$. As a result, the minimum inductance occurs at zero-current, around which $L(I)$ appears to be symmetric. On the other hand, in the presence of an in-plane magnetic field component along y-direction ($B_y \neq 0$), CPR becomes asymmetric around inflection-point $(i^*,\varphi^*)$, mainly associated with the broken Kramers degeneracy between the oppositely polarized spin components of Andreev bound states (ABS) leading to a finite-momentum pairing. As a result, the current dependence of the Josephson inductance $L(I)$ also become asymmetric and the minimum of $L(I)$ appears at some finite current $i^*$, corresponding to the shifted inflection point $(i^*,\varphi^*)$ in the CPR. Such a pronounced asymmetry in the skewed CPR and, thus, in the Josephson inductance L(I), signals the supercurrent MCA (as defined in equation (4)) and hence supercurrent SDE. Further details on inductive measurements and the nonreciprocity in the Josephson inductance/current can be found in the Ref.[27], where it is explicitly observed that the value of $\gamma_L$, obtained from inductive measurements performed at T = 100 mK, far below the transition temperature ($T_c \sim 1.45$ K), is of the same order as that of $\gamma_S$ calculated for resistive measurements at $T_c$.

## Fulde–Ferrell–Larkin–Ovchinnikov state

In the field of conventional superconductivity, following from the fact that Cooper pairing is formed between Kramers partners and the most known conventional superconductors are characterized by the Bardeen–Cooper–Schrieffer (BCS) theory[43], the presence of time-reversal symmetry is a key ingredient and the preserved Kramers degeneracy is the fundamental reason/criterion that stabilizes superconducting phase in so many systems at sufficiently low temperatures[44–46]. Thus, due to electron pair breaking, such a conventional superconducting state with a spin-singlet pairing is suppressed or destroyed by time-reversal symmetry breaking perturbations ¯ as a consequence of applied magnetic field, doped magnetic impurities, proximity-induced magnetization, or intrinsic magnetic instability leading to spontaneous magnetization.

On the other hand, beyond the conventional BCS paradigm, unconventional superconductivity allows coexistence of more exotic superconducting order parameters with magnetism. For instance, as predicted independently by Peter Fulde and Richard Ferrell (FF)[47] and Anatoly Larkin and Yuri Ovchinnikov (LO)[48], magnetic fields can give rise to a superconducting state with FF-type order parameter $\Delta(x) = \Delta e^{iqx}$ and/or spatially inhomogeneous LO-type pair potential $\Delta(x) = \Delta\cos qx$. The underlying physical mechanism of the Fulde–Ferrell–Larkin–Ovchinnikov (FFLO) state[47,48], owning to the opposite energy-shift in the electronic spin bands, induces non-zero centre-of-mass momentum of Cooper pairs and leads to a spatially-modulated order parameter. The FF state ubiquitously exists in noncentrosymmetric superconductors and is particularly known as the helical superconductivity[49–61].

The FFLO states, and/or the implications of the helical superconductivity, have been obtained in heavy-fermion superconductors $CeCoIn_5$[62–64], organic superconductors[65], pure single crystals of FeSe[66,67], thin films of Pb[60] and doped $SrTiO_3$[68], a heavy-fermion Kondo superlattice[69,70], and a three-dimensional topological insulator $Bi_2Se_3$[71]. While the existence of FFLO-like states is well-established in proximity-coupled superconductors and ferromagnets[72], the experimental observation of FFLO states has been reported in nonmagnetic superconductors by applying external magnetic fields[62,63] as well as intrinsic ferromagnetic superconductors[73–79].

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