Diode effect in superconducting Nb$_3$Sn micro-bridges at high frequencies

Sara Chahid, Serafim Teknowijoyo, Iris Mowgood, and Armen Gulian

Advanced Physics Laboratory, Institute for Quantum Studies, Chapman University, Burtonsville, MD 20866, USA

(Dated: November 22, 2022)

The superconducting diode effect has been recently reported in a variety of systems. Various symmetry breaking mechanisms have been examined. However, frequency ranges of these potentially important devices are still obscure. We investigated superconducting micro-bridges of Nb$_3$Sn with a relatively large diode efficiency of $\sim 5\%$ using an out-of-plane magnetic field as small as 2.5 mT; optimum magnetic fields (up to 10 mT) generate higher efficiency (up to 10\%) while higher fields, $\sim 15-20$ mT, quench the effect. The diode changes its polarity with the magnetic field. Interestingly, the bridge resistance at diode operation reaches a value with a factor of 2 times smaller than in its normal state, which is compatible with the vortex-caused mechanism of resistivity. Further confirmation of this mechanism was revealed by scanning electron microscopy: dissimilar edges of the superconductor strip can be responsible for the inversion symmetry breaking. Superconductive diode rectification was observed at frequencies up to 100 kHz, the highest reported as of today. Estimates are in favor of much higher, GHz, range of frequencies.

I. INTRODUCTION

For certain important problems of fundamental physics, for example, exploration of quasi-local action of curl-less vector potential [1], it is necessary to use superconducting micro-bridges for which the magnitudes of critical currents of superconductor bridge are equal for both polarities. However, because of simultaneous breaking of time reversal symmetry (TRS) and inversion symmetry (IS), the phenomenon called the superconducting diode effect (SDE) arises which violates this equality [2, 9]. The latest research focus in superconductivity has been this effect with a potential of being applied in significantly less-dissipative superconducting electronics. In theoretical models, the TRS is broken by externally applied magnetic field or internal inclusions of magnetic micro-clusters, while the IS is broken by the out-of-plane Rashba spin–orbit coupling [10–13], valley-Zeeman interaction [14], etc., which results in the emergence of a chiral superconducting order [10, 12–14]. Experimentally, systems based on van der Waals material MoS$_2$ with non-centrosymmetric crystal potential [15], synthetic super lattice of Nb/V/Ta [5], and some others [17] have been reported as well as planar Josephson junction arrays of Al on InAs [1, 18]. Yet some other systems reveal nonreciprocal behavior in field-free environments, such as NbSe$_2$-based Josephson junction [8] and tri-layer graphene [19]. In view of variety of experimental observations and theories on SDE in different configuration of systems, it is quite reasonable to assume that more than one mechanism can be responsible for the ubiquity of nonreciprocity observations in superconductor thin films [20]. In a recent article [21], the SDE was found in NbN micro-bridges in an out-of-plane magnetic field. This observation was attributed to the critical current being determined by the vortex flow, confirming that the SDE is caused by unequal vortex barriers on the two edges of the bridge [22, 23].

This mechanism is easy to understand. Consider a type-II superconductor film strip. A magnetic field above $H_{c1}$ creates vortices; initially, they nucleate at the strip edges [24, 25]. Morphology of these edges affects the surface barrier, which prevents the vortices to enter into the strip [26, 27]. However, if the current through the strip is strong enough, the Lorentz force which it exerts onto the vortex overcomes the surface barrier; vortices start moving across the strip, thus dissipating energy and creating a resistive state. In practice, vortex barriers are unequal because of non-identical nature of strip edges. This circumstance has been used by Vodolazov and Peeters [23] at predicting superconducting diode effect in 2005 and experimentally confirmed recently [21].

The spike of activities in the area of superconducting diodes is paving a road towards future practical application of these novel devices in superconducting electronics. However, a very important topic is still open and remains unaddressed: that is the frequency range of SDE. No reports on the frequency range above few hertz can be found in the literature which spreads doubts in practical applicability of the devices. To address this topic, we explore SDE up to 100 kHz frequencies, and express arguments that the frequency range can be even orders of magnitude higher. This is the main result of our report.

II. EXPERIMENTAL DETAILS

Our study used conventional Nb$_3$Sn superconducting thin film bridges. The Nb$_3$Sn films were prepared in a DC/RF magnetron sputtering system (manufactured by AJA International, Inc.) with a base pressure of $1 \times 10^{-8}$ Torr. The Nb target (Kurt Lesker, 99.95\%) was placed inside a DC gun while the Sn target (Kurt Lesker, 99.999\%) was sputtered using an RF source to avoid melting. The sapphire substrate (AdValue Technology,
thickness 650 µm, C-cut) was cleaned thoroughly with isopropyl alcohol before it was mounted on the holder. In our chamber’s configuration, the substrate holder is at the center of the chamber facing upwards, while the sputtering guns (up to 5) are located at the top. The substrate is rotated in-plane throughout the whole deposition process to ensure homogeneous deposition layer over the whole surface. Our pre-deposition in-situ cleaning of the substrate involves heating it up to 900 C for 10 min then followed by a gentle bombardment of Ar+ ions at 400 C for 5 min. The substrate was oriented to face the ion gun squarely. For the Nb3Sn film deposition, we used a “stack+anneal” process similar to the one in [28]. Taking advantage of Nb–Sn phase diagram [29], an alternating Nb and Sn stacks with an excess Sn content (Nb:Sn ratio ~ 2) “phase-locks” into the Nb3Sn composition when the excess Sn evaporates during post-deposition annealing. For the first/base layer, Nb was sputtered in DC mode with 500 W power and 3 mTorr pressure at 600 C for 20 min. Then Sn layer was deposited on top of Nb in RF mode with 250 W power and 30 mTorr pressure at 100 C for 15 min. Nb was deposited one more time as the capping (third) layer using the same parameters as the base layer for 3 min. Finally, the substrate is in-situ heated to 950 C for 30 minutes to let the excess Sn evaporate before cooled down to room temperature. All the heating/cooling protocols always used a 30 C/min ramp rate. Our films are ~ 100 nm thick and resistivity measurements in PPMS (Quantum Design) confirmed its stoichiometry and homogeneity from their consistent Tc ~ 17.6 K and RRR(300K/20K) ~ 4 between samples across several batches; the stoichiometry was also checked by the SEM EDX (Hitachi SU3500, Oxford Instruments X-MAX-20).

Figure 1 demonstrates certain physical properties of our films (panels a and b) and bridges (panels c-f). Patterning of the films into bridges has been performed using combination of 3D−printing (Elegoo Mars-3 printer), photolithography and by ion milling with macroscopic contact pads on 1 × 1 cm² sapphire substrates. Reactive ion milling (Bal-Tec RES-101, CF4 etchant) was used to develop metallic film pattern, Fig. FIG. 1(c). After removal of the resin, the structure was covered by negative photoresist, and the 100 µm-scale bridge was narrowed down to 12 µm with the projective photolithography (using mask projection via LUMAM epi-fluorescent microscope, 10× objective). After ion milling, the last stage of patterning was undertaken using positive photoresist and the same projective technique with the 40× objective and a different mask in the form of two holes. The final ion milling delivered bridges about 10 µm long and down to 2 µm wide, Fig. FIG. 1(d).
FIG. 2. (a) Resistive state at various values of bias temperature for the applied current of opposite polarities. (b) SDE with sinusoidal current amplitude $\approx 10$ mA and frequency 0.1 Hz in 2 $\mu$m bridge. (c) Same as in panel (b) with 5 $\mu$m-wide bridge at frequency 10 kHz. (d) SDE at frequency 100 kHz (average of 200 acquisitions) at reversed polarity of magnetic field. In all cases, a magnetic field orthogonal to the surface of the bridge was applied and optimized in the range 50-100 Oe.

Ion milling affects the physical properties of bridges thus reducing the critical temperatures down to the 2 – 12 K range. This $T_c$-reduction was also noticed in the literature [21]. We were able to restore the $T_c \sim 17$ K values in some of the bridges by high-vacuum post-annealing ($\sim 10^{-6}$ Torr) at 900 C during 30 min, as shown in Fig. 1(e).

III. RESULTS

The tests demonstrated $V(I_+ \neq V(I_-)$, Fig. 1(f), thus indicating SDE. To study the SDE, temperature dependence of 2 $\mu$m bridge was measured, Fig. 2(a).

This measurement confirms the fact mentioned in literature that at lower temperatures the difference between the threshold values of $I_{res}^+$ and $I_{res}^-$ increases with the decreasing bias temperature [5, 20] (the value of $I_{res}^+$ corresponds to the current at which the resistive state sets up). The bridge then was biased at 2 K temperature (PPMS DynaCool cryostat) and the expected diode effect was confirmed, Fig. 2(b), with externally applied current source and nanovoltmeter (Keithley 6221 and 2182a respectively) at frequency 0.1 Hz.

Since superconducting diodes are considered as important elements for electronics, it was meaningful to register the effect at possibly higher frequencies. The Keithley current source generates AC-currents up to 100 kHz. Detection of voltage output was performed by oscilloscope (Tektronix TDS 644A). The results are shown at Fig. 2(c) and (d) for 10 kHz and 100 kHz frequencies, respectively. Higher frequency measurements require specially designed circuitry in the cryostat since the noise becomes an issue, as is visible from the comparison of Figs. 2(d) and (b). For reducing noise, we plotted 100 kHz data averaged over 200 acquisitions.

IV. DISCUSSION

To estimate the frequency range of this type of diodes, one needs to understand what is the underlying mechanism of the SDE. Comparison of panels (a) and (b) in Fig. 2 shows the diode polarity change because of switching the external magnetic field from 100 Oe to $-100$ Oe. The field is orthogonal to the bridge surface. The field amplitude is optimal for SDE in our bridges. Higher values ($\sim 150$ Oe) quench the effect. The resistive state which yields the non-zero voltage in Fig. 2(a) is caused by the vortex motion: the correspond-
FIG. 3. $V(I)$ dependence of 2 µm bridge. Resistive state jumps are noticeable before the transition to the normal resistive state.

ing value of resistance is about the half of its normal value. Indeed, as follows from Fig. 3, the resistance ($V/I$) reaches the value of 25 – 40 Ohm, while in normal state, just above the superconducting transition, the resistance is $\sim$ 60 Ohm. Figure 3 depicts a set of $V(I)$ measurements at various temperatures. Multiple resistivity jumps are noticeable at lower temperature curves. This kind of the $V(I)$ curves behavior is in close correspondence with the results for Abrikosov and kinematic vortices and phase-slip lines revealed by modeling in thin-film bridges [21, 30–32]. As was shown in via modeling in these works, different values of the applied current correspond to different vortex lattice patterns. We reproduced the computations by Vodolazov and Peeters [32] using time-dependent Ginzburg-Landau (TDGL) equations and COMSOL Multiphysics package in a slightly different model: our bridge has geometrically broken IS: its top edge has three shallow dents (with depth less than the vortex lateral size), while the bottom edge is smooth. Such a model allows us not only explain the $V(I)$ characteristics in Fig. 3 similarly to [32], but also certain diode features.

We will accentuate on the latter ones here. Figure 4 demonstrates the vortex lattice pattern in the given external magnetic field $H < H_{c2}$ at both polarities of applied current.

As follows from this figure, the evolution is essentially different due to the presence of (three) small dents in the top edge of the bridges. It breaks the IC symmetry, and the evolution of the system differs for opposite polarities of the current. At large enough values of the current, the individual vortices are consolidating into lines crossing the width of the bridge. They are classified as Phase Slip Lines (PSL). Again, the presence of minor dents creates asymmetry between the opposite current polarities: to generate the full width crossing by PSL the positive polarity current required 50% longer time than the negative current.

In case of our experiments, the IS breaking, which is the second required condition for SDE, is being provided, most likely, by the breaking of left-right edge symmetry in lithographically processed bridges. Closer look to Fig. 1 (d, Inset) illustrates the fractured structure of the edge of the current flow channel.

Associating the SDE with the vortex motion, we can make certain estimates for the performance limits of SDE in the frequency domain. The 100 kHz results have been obtained in 5 µm-wide bridge. One can relate this frequency with the reciprocal characteristic vortex crossing time of the bridge. If the geometry is scaled down to 5 – 50 nm-wide bridges, it can deliver 2 – 3 orders of magnitude higher frequencies because of geometric factors. Can even higher frequency range be reached? The answer looks positive because of the following rea-
Ordinary thin superconducting bridges (in our case, Nb$_3$Sn) can demonstrate SDE because of simultaneous breaking of IS and TRS. Natural reasons of breaking the IS is the geometric asymmetry between the bridge edges, and the TRS can be broken by the remnant magnetic field in the cryostat or by the current via the bridge itself. Finite element modeling of physical phenomena in the bridge based on the TDGL equations reveal complex microscopic nature of the resistive states in it. More experiments guided by the results of this modeling can yield higher-performance two terminal and multi-terminal devices useful for practice.

ACKNOWLEDGMENTS

The work of Chapman U. research team is supported by the ONR grants N00014-21-1-2879 and N00014-20-1-2442.
uitous Superconducting Diode Effect in Superconductor Thin Films, arXiv.2205.09276 (2022).

[21] D. Suri, A. Kamra, T. N. G. Meier, M. Kronseder, W. Belzig, C. H. Back, and C. Strunk, Non-reciprocity of vortex-limited critical current in conventional superconducting micro-bridges, Applied Physics Letters 121, 102601 (2022).

[22] M. K. Hope, M. Amundsen, D. Suri, J. S. Moodera, and A. Kamra, Interfacial control of vortex-limited critical current in type-II superconductor films, Phys. Rev. B 104, 184512 (2021).

[23] D. Y. Vodolazov and F. M. Peeters, Superconducting rectifier based on the asymmetric surface barrier effect, Phys. Rev. B 72, 172508 (2005).

[24] V. V. Shmidt, The critical current in superconducting films, Sov. Phys. JETP 30, 1137 (1970).

[25] V. V. Shmidt, Critical currents in superconductors, Sov. Phys. Usp. 13, 408 (1970).

[26] C. P. Bean and J. D. Livingston, Surface Barrier in Type-II Superconductors, Phys. Rev. Lett. 12, 14 (1964).

[27] K. Fossheim and A. Sudboe, Superconductivity: Physics and Applications (Wiley, 2004) p. 232.

[28] C. S. Sundahl, Synthesis of Superconducting Nb3Sn Thin Film Heterostructures for the Study of High-Energy RF Physics, Ph.D. thesis, The University of Wisconsin - Madison, ProQuest Dissertations Publishing, 13805402 (2019).

[29] J. P. Charlesworth, I. Macphail, and P. E. Madsen, Experimental work on the niobium-tin constitution diagram and related studies, Journal of Materials Science 5, 580 (1970).

[30] G. R. Berdiyorov, A. K. Elmurodov, F. M. Peeters, and D. Y. Vodolazov, Finite-size effect on the resistive state in a mesoscopic type-II superconducting stripe, Phys. Rev. B 79, 174506 (2009).

[31] P. Sánchez-Lotero, J. Albino Aguiar, and D. Domínguez, Behavior of the flux-flow resistivity in mesoscopic superconductors, Physica C: Superconductivity and its Applications 503, 120 (2014).

[32] D. Y. Vodolazov and F. M. Peeters, Rearrangement of the vortex lattice due to instabilities of vortex flow, Phys. Rev. B 76, 014521 (2007).