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Abstract

In this Perspective article, we evaluate the current state of research on the use of focused electron and ion beams to directly fabricate nanoscale superconducting devices with application in quantum technologies. First, the article introduces the main superconducting devices and their fabrication by means of standard lithography techniques such as optical lithography and electron beam lithography. Then, focused ion beam patterning of superconductors through milling or irradiation is shown, as well as the growth of superconducting devices by means of focused electron and ion beam induced deposition. We suggest that the key benefits of these resist-free direct-growth techniques for quantum technologies include the ability to make electrical nanocontacts and circuit edit, fabrication of high-resolution superconducting resonators, creation of Josephson junctions and superconducting quantum interference device (SQUIDs) for on-tip sensors, patterning of high-Tc SQUIDs and other superconducting circuits, and the exploration of fluxtronics and topological superconductivity.

1. Superconducting devices in quantum technologies and their fabrication

Superconductivity, discovered in 1911 by H K Onnes, is a relative rare emergent phenomenon, quantum in nature and with macrocopic implications such as the lack of electrical resistance to the passage of electrical current [1]. Superconductors, beyond representing an excellent playground to investigate fundamental physical phenomena [2], are crucial materials in today's and future's technology [3, 4]. In quantum technologies, superconductors have found applications in quantum computing and in quantum sensing. At the very heart of quantum computing, one can find Josephson junctions (JJs), in which two close superconductors (S) are separated either by an insulator (I), SIS, a normal metal (N), SNS, or a different superconductor (s), SsS [5, 6]. Described for the first time by B D Josephson in 1962, JJs display a rich variety of physical phenomena, with the existence of an electrical supercurrent as one of the key features [7]. Central to the interest of superconductors for quantum technologies is the fact that the ground state of a superconductor is a coherent state with a single phase [8]. In the BCS theory of superconductivity, the electrical current is carried by Cooper pairs that condense into the same ground (quantum) state. Cooper pairs can flow through a JJ, and the electrical behaviour of the JJ is determined by the value of the superconducting phase on both sides of the junction. In addition, a JJ has a strong interaction with microwave radiation [7, 9]. In the scientific literature, one can find a rich variety of quantum devices based on superconductors, which can be classified according to their final application into (a) superconducting circuits for quantum computing and (b) quantum sensors based on superconductors.

In the (a) category, JJs contribute to realize qubits thanks to the fact that the JJs endow the circuit with non-linearity without introducing dissipation or dephasing [10–12]. The non-linearity leads to non-equidistant energy levels, as required for qubits. Various practical implementions of superconducting-based qubits have been built, including the transmon, the fluxonium, the quantronium and the hybrid qubits [12, 13]. Besides, in these and other qubit architectures, superconducting materials can provide key-enabling circuit features such as the realization of high-frequency resonators and bias lines [14, 15]. Moreover, proposals of Majorana-based quantum computing based on topological superconductivity induced by proximity effects have been put forward [16, 17]. On the other hand, an intense research activity is under way to overcome the materials challenges existing for the realization of practical quantum computers [18]. In the (b) category, one can find superconducting sensors such as superconducting quantum interference device (SQUIDs) [19–21], transition-edge detectors [22, 23], nanowire single-photon detectors [24, 25] and kinetic inductance detectors [26].

The fabrication of superconducting devices call for the use of micro- and nano-fabrication techniques, leveraged from their use in semiconductor industry or in micro/nano-technology. In general, these techniques are based on top-down multi-step lithography processes that can be additive or subtractive and are used in combination with thin-film deposition techniques. Aluminum (Al) and niobium (Nb) are frequently used as superconducting materials due to their good behaviour at the typical working temperature, in the mK range. The technology of fabrication of JJs based on these two materials is mature, which facilitates its application to circuits designed for quantum computing [27, 28]. However, defects at the surface of the devices or on the substrate are important sources of decoherence [29–32]. For quantum sensors and key-enabling features, there is a broader choice of superconducting materials beyond Al and Nb. One of the most popular superconducting materials is YBCO ($YBa_2Cu_3O_{7-x}$), which stands out for its high T_C (∼90 K) and allows for JJs and SQUIDs up to temperatures close to T_C [19]. The patterning of exotic superconductors such as LAO/STO interfaces [33] and proximitized graphene [34] into JJs has also been achieved, as well as JJs and SQUIDs based on proximitized topological insulators [35].

For the fabrication of these superconducting devices, various lithographic techniques have been successfully applied. A few representative examples can be cited, without pretending to cover an immense topic that is not the main focus of the current article:

- Photolithography has been used for integrating superconducting routing levels on a Si-based interposer with the aim of electrically coupling spin qubit arrays with cryo-CMOS control and read-out circuits [36].
- Electron beam lithography is a very popular fabrication technique in quantum technologies and has been applied to fabricate Josephson persistent-current qubits [37], flux qubits [38], phase qubits [39], etc. The Sycamore processor, based on an array of transmon qubits, has been fabricated by means of 14 lithography layers utilizing both, optical and electron beam lithography [28].
- Stencil mask plus shadow evaporation technique has been used to fabricate artificial topological superconductors showing Majorana bound states [40].
- Direct-write techniques have been utilized in various ways for quantum technologies and will be the focus of the next section, which constitutes the central discussion point of the current perspective article.

2. Nanoscale direct-write fabrication of superconducting devices

In sharp contrast to optical or electron beam lithography, nanoscale direct-write fabrication techniques do not require the use of sacrificial resists or etching/lift-off steps, avoiding contamination issues arising from organic residues. The level of maturity reached by optical and electron beam lithography is higher than that of direct-write fabrication, which justifies their preferred use at this moment. However, in some applications requiring nanoscale superconductors, standard optical or electron beam lithography techniques may not meet the requirements in terms of resolution, substrate compatibility, geometry, etc, which opens up the possibility of using alternative nanofabrication strategies based on focused electron and ion beams. In the following, we will examine some examples published in recent years where these alternative techniques have been successfully applied.

2.1. Focused ion beam (FIB) patterning of superconductors

FIB is well suited for high-resolution removal of material through local sputtering, which has been widely applied to pattern superconductors and fabricate various types of superconducting micro/nano-structures and devices. The following strategies can be found in literature:

- By means of Ga^+-FIB milling on superconducting MgB_2 with $T_C \sim 40$ K [41], planar JJ [42–44] as well as SQUIDs [45] were fabricated. JJ patterning was also achieved by Ga^+-FIB milling of high-T_C superconductors such as BISCCO [46] and YBCO [47–50]. On YBCO, it is possible to use Ga^+-FIB milling to create nanowires [51] and sub-micron features [52] as well as full SQUID nanodevices [53].
- In a different approach, Ga^+-FIB patterning of single crystals has been achieved, allowing the investigation of anisotropic superconducting properties in SmFeAs(F, O), with $T_C = 54$ K [54], the spontaneous emergence of JJs in homogeneous rings of single-crystal Sr_2RuO_4 [55] and tailored superconducting landscapes [56].

- In the quest for advanced scanning-SQUID sensors, Ga^+-FIB patterning of a thin Nb layer grown on a cantilever has been performed. The resulting SQUID-on-tip (SOT) sensor has been used to image small magnetic signals with high lateral resolution [57]. Moreover, such SOT sensors have been applied to image the magnetic flux arising from a superconducting qubit [58].
- Superconducting microwave resonators in the proximity of qubits are of great interest to control them, which can be fabricated by Ga^+-FIB [59, 60] as well as by Ne^+-FIB with a better resolution [61]. For milling purposes, Ga^+-FIB is capable of higher throughput compared to Ne^+-FIB and He^+-FIB, but the use of these light ions is preferred for high resolution given the smaller ion beam spot and the weaker interaction with the material to mill [62].

Remarkably, He^+-FIB irradiation using low doses have been used to create planar JJs and SQUIDs on YBCO [63]. The procedure allows reaching 10 nm resolution in the patterning of JJs [64, 65]. Using this technology, a low-power high-T_C superconducting single flux quantum circuit has been achieved [66], with potential applications for memory, mixers, digital sensor readouts, switches for routers, and high-speed computing.

2.2. Direct growth of superconducting devices by focused electron and ion beam induced deposition (FEBID and FIBID)

FEBID and FIBID respectively, are direct-write resist-free nanolithography techniques that enable the growth of high-resolution nano- and micro-structures [67, 68]. They rely on a gas precursor that is injected into the area of interest and decomposed by a focused electron or ion beam, giving rise to deposits exhibiting functional properties such as tunable electrical conductivity (insulating, semiconducting or conducting behaviour) [69, 70], ferromagnetism [71, 72], nano-optical or photonic behaviour [73, 74], superconductivity [75, 76], robust structural and mechanical properties [77, 78], etc.

Since 2004, when superconductivity ($T_C = 5.2$ K) was discovered in W-C deposits by FIBID using the $W(CO)_6$ precursor [75], a high number of publications have reported the use of FEBID and FIBID for growing superconducting structures, as recently reviewed in detail by our group [79]. The reader is invited to read that publication in order to find all types of reports on the topic, whereas here, we will focus on applications of superconducting deposits by FEBID and FIBID in the field of quantum technologies. In the following, we will highlight a few results within this domain:

- On W-C nanowires grown by FIBID, magnetotransport results below T_C have been interpreted as due to the presence of phase slips and quantum phase transitions [80]. Interestingly, a laterally-applied gate voltage in W-C nanowires allows tuning a superconducting to normal-state transition [81].
- In the arena of fluxtronics, a rich and far-reaching literature has reported the behaviour of superconducting vortices on W-C FIBID deposits. Whereas the first publications focused on the properties of the Abrikosov vortex lattice against changes in temperature and magnetic field [82, 83] and the effects caused by the built-in disorder [84–86], subsequent studies addressed the formation of a single row of vortices in ultra-narrow W-C nanowires [87] and its manipulation by means of Lorentz forces [88, 89], including the discovery of ultra-fast vortex movement on Nb-C nanowires [90].
- More recently, the fabrication of JJs and SQUIDs based on FEBID and FIBID deposits has been achieved. By using W-C deposits grown by FEBID under high and low electron current, superconducting electrodes connected by normal electrodes have been created, with Shapiro steps confirming the formation of JJs [91]. Also, arrays of Nb-C JJs have been deposited by FIBID [92]. In addition, nanoSQUIDs based on W-C deposits grown by FIBID have been fabricated with Dayem-bridge-type geometry [93]. Such nanoSQUIDs are expected to be directly grown on cantilevers for scanning-on-tip devices, which would allow sensing small magnetic fields, currents and dissipation with high lateral resolution, as mentioned in the previous section [57]. Interestingly, W-C deposits have been applied to the editing of superconducting circuits based on other superconductors [94].
- W-C FIBID deposits have been frequently used to induce superconductivity by proximity effect on other materials such as graphene [95], bismuth [96, 97], copper and cobalt [98], gold [99] and Bi_2Se_3 [100]. In the last case, topological superconductivity was searched for Majorana-based quantum computing applications. In this topic, it is interesting to note that the critical magnetic field B_{C2} is very high in W-C FIBID deposits, with reports showing values up to 11 T [101].
- FEBID and FIBID excel in the growth of three-dimensional nanostructures [102, 103], which in general is not possible with other nanolithography techniques. In this topic, our group has successfully shown that superconducting W-C deposits with nanotube [104] and nanohelix [105] geometries can be grown, leading to a noticeable interplay of the vortex lattice with these unconventional geometries. In this regard, a perspective article has been recently published on superconductivity in curved 3D nanoarchitectures [106].

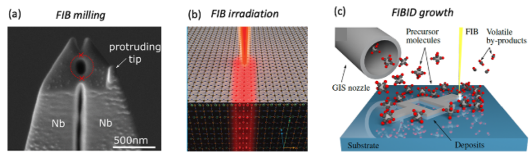

Figure 1. (a) NanoSQUID fabricated on a commercial AFM cantilever through FIB milling of a deposited 50 nm Nb film. Reprinted figure with permission from [57], Copyright (2022) by the American Physical Society. (b) Sketch illustrating the local irradiation of a high-T_C YBCO film by means of a He^+-FIB in order to directly create a JJ. Reproduced from [63], with permission from Springer Nature. (c) Sketch of the direct growth of a nanoSQUID using the FIBID technique. A detailed description of the process has been published in Sigloch *et al*, Nanoscale Advances 4, 4628 (2022) [93].

In figure 1, three of the devices discussed in section 2 are shown as examples of the current capabilities of direct-write fabrication of superconducting devices for quantum technologies.

3. Outlook

Optical and electron beam lithography techniques are mature techniques for the fabrication of reproducible and reliable Al-based and Nb-based superconducting devices applied in quantum technologies. Despite these being the preferred lithography techniques and the preferred superconducting materials in this domain, the use of focused electron and ion beams for nanoscale direct-write fabrication of superconducting devices is nevertheless increasing. In this regard, several strategies can be used, such as FIB milling, FIB irradiation and FEBID/FIBID growth of superconductors. This set of techniques (FIB, FEBID, FIBID) is particularly well suited for obtaining devices with high lateral resolution and for building devices that rely on unusual geometries and unconventional substrates. Thus, they represent a convenient approach to create a nanopatterned superconducting material on a targeted location without the burden of conventional multi-step processes and avoiding the residues caused by the use of sacrificial resists. On the other hand, ion implantation, especially if Ga^+-FIB is used, can be disadvantageous when used on particular materials [107]. Although these techniques present an intrinsic difficulty for wafer-scale patterning given the slow movement of the ion and electron beams, in the current state of the quantum technology some opportunities exist. Here below, we summarize some niches within the field of quantum technologies in which direct patterning by focused electron and ion beams could make an impact, as well as some of the foreseeable challenges that will be encountered.

(1) *Electrical nanocontacting and circuit edit*. Similarly to the case of semiconductor industry [108, 109], the direct use of focused electron and ion beams to remove or to add materials locally is applicable to create or reconfigure electrical connections in a superconducting circuit. The fact that superconducting materials themselves can be directly grown by FEBID/FIBID represents an added value. Challenges here are the relatively high electrical resistance of the grown materials and the reproducibility of the composition and physical properties of the deposited materials [79].
(2) *High-resolution superconducting resonators*. FIB milling is appropriate to define constriction-type weak links on superconductors for frequency-tunable magnetic-field-resilient high-quality resonators in the microwave range, which are of interest in hybrid superconducting-spin systems [59, 61]. The limited milling resolution provided by Ga^+ FIB sources can be overcome by the use of He^+–Ne^+ FIB sources and other new ion sources. It is nevertheless foreseeable that commercial quantum devices based on this hybrid approach would call for a wafer-scale process being fully compatible with optical and/or electron beam lithography.
(3) *JJs and SQUIDs for on-tip sensors*. FIB-based milling [57], irradiation [63] and deposition [93] seem convenient to create superconducting sensors on tips and cantilevers, in particular JJs and SQUIDs, with the potential to investigate relevant physical properties of materials and devices at the nanoscale [58]. These specialized sensors could represent a niche for FIB-based processing given that the use of standard lithography might be possible here, but difficult for up-scaling [110, 111]. However, further developments are still needed to create a marketable product with sufficient number of end users.
(4) *High-Tc SQUIDs and other superconducting circuits*. Whereas the use of very low temperatures is recommended for quantum computing in order to minimize decoherence, other applications such as sensing do not have this limitation. In this latter case, YBCO-based SQUIDs patterned either by FIB milling [53] or by FIB irradiation [65] have been demonstrated. Moreover, the reported high-Tc superconducting single flux quantum circuit is promising for quantum logic above liquid helium

temperature [66]. However, this approach is restricted to a few research groups and still far from the maturity and fabrication throughput required by industry.

(5) *Fluxtronics*. The fact that a superconducting vortex embodies a single flux quantum and behaves as a particle when submitted to an electric current opens the way for sensing and logic applications based on vortices [112, 113]. FIB-based devices have demonstrated their potential in this field, including the long-range (10 μm) coherent vortex transfer [88], the ultra-fast vortex displacement [90] and the fabrication of 3D fluxtronic circuits [105], but further work is needed to substantiate the true competitiveness of fluxtronics in quantum applications.

(6) *Topological superconductivity*. With topological superconductivity being a holy grail for Majorana-based quantum computing [16, 17], the use of FEBID/FIBID-grown superconducting deposits capable of inducing local superconductivity by proximity effect is valuable, as already demonstrated in literature [96, 97]. On the other hand, FEBID/FIBID techniques are not appropriate to achieve high-quality epitaxial interfaces between semiconductor nanowires and superconductors, as standard lithography techniques have recently demonstrated [114].

As a concluding remark, let us suggest that we consider direct-write techniques such as FIB milling, FIB irradiation and FEBID/FIBID valuable for quantum technologies in the following domains: prototyping (*electrical nanocontacting and circuit edit, high-resolution superconducting resonators, high-Tc SQUIDs and other superconducting circuits*), exploration of new effects (in *fluxtronics* and in *topological superconductivity*) and niche fabrication (*JJs and SQUIDs for on-tip sensors*).

Data availability statement

No new data were created or analysed in this study.

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