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Control of bulk superconductivity via surface-bound electric fields in ion-gated niobium nitride thin films

E. Piatti[1], D. Daghero[1], G. A. Ummarino[1], M. Colangelo[2], D. Romanin[1], O. Medeiros[2], F. Galanti[1], F. Laviano[1], J. R. Nair[1], A. Sola[3], C. Portesi[3], R. Cristiano[4], A. Casaburi[5], I. Yu. Sklyadneva[6], E. V. Chulkov[6], R. Heid[7], K. K. Berggren[2], and R. S. Gonnelli[1]

[1] Department of Applied Science and Technology, Politecnico di Torino, Torino, Italy
[2] Department of Electrical Engineering and Computer Science, Massachusetts Institute of Technology, Cambridge, USA
[3] INRIM – Istituto Nazionale di Ricerca Metrologica, Torino, Italy
[3] CNR-SPIN Institute of Superconductors, Innovative Materials and Devices, Napoli, Italy
[5] School of Engineering, University of Glasgow, Glasgow, United Kingdom
[6] Donostia International Physics Center, San Sebastian, Basque Country, Spain
[7] Karlsruher Institut für Technologie, Institut für Festkörperphysik, Karlsruhe, Germany

The electric field effect is the modulation of the electronic properties of a material by the application of a transverse electric field. Its most influential application is the field-effect transistor (FET), which forms the backbone of modern semiconductor electronics. The possibility to control the superconducting (SC) state by means of the electric field effect has been an open issue in condensed matter physics since the invention of the FET in 1960. Early attempts included the experiment of Glover and Sherril [1] where they replaced one electrode of a capacitor with tin and indium SC films, obtaining a faint 0.002% shift in the original SC transition temperature $T_c = 3.72$ K; and the one of Stadler [2], where by substituting the solid-oxide dielectric with a ferroelectric the T_c shift was enhanced to 0.035%. These seminal results confirmed both the feasibility of controlling the SC phase via the electric field effect, as well as its negligible impact for device applications, and were promptly forgotten for almost forty years. The discovery of high-T_c superconductors lead to a rekindled interest in the topic, since the much smaller carrier density of the cuprates with respect to metallic superconductors allowed a much more effective tunability. Examples included an electric-field-induced 10 K T_c shift in YBCO [3], and full insulator-to-superconductor transitions in ultrathin Bi [4] and GBCO [5], but still the range of applicability remained limited.

The turning point came in the late 2000s with the introduction of the ionic gating technique, which rapidly became a very popular tool to investigate and control the electric transport and electronic ground state in a wide variety of different materials. Ionic gating operates by exploiting the ultrahigh electric fields that build up at a voltage-polarized solid-electrolyte interface in the so-called electric double layer (EDL), made possible by the sub-nanometric spacing between the two charged layers in the EDL. When incorporated in an EDL-FET architecture, this allows attaining modulations of the surface charge density in the device channel which are often comparable to those occurring in metallic systems. Since then, the ionic gating technique has found large success in tuning the phase diagram of low- and moderate-carrier density systems, including cuprates and iron-based superconductors. Nevertheless, its applicability to conventional metallic superconductors has received significantly less attention, and – excluding the work carried out by my research group – investigations have been restricted to niobium thin films, where T_c shifts of 0.9% were observed [6].

In this Invited Talk, I will present the work which has been carried out over several years by my research group to investigate how ionic gating can tune the properties of metallic superconductor, using niobium nitride (NbN) as an emblematic case [7]. We selected NbN due to it being a standard example of electron-phonon-driven metallic superconductor, it being electrochemically stable and mechanically robust, and because preliminary DFT calculations indicated that its T_c could be nearly doubled if a sufficiently large number of electrons were to be added to its first unit cell from the surface [8].

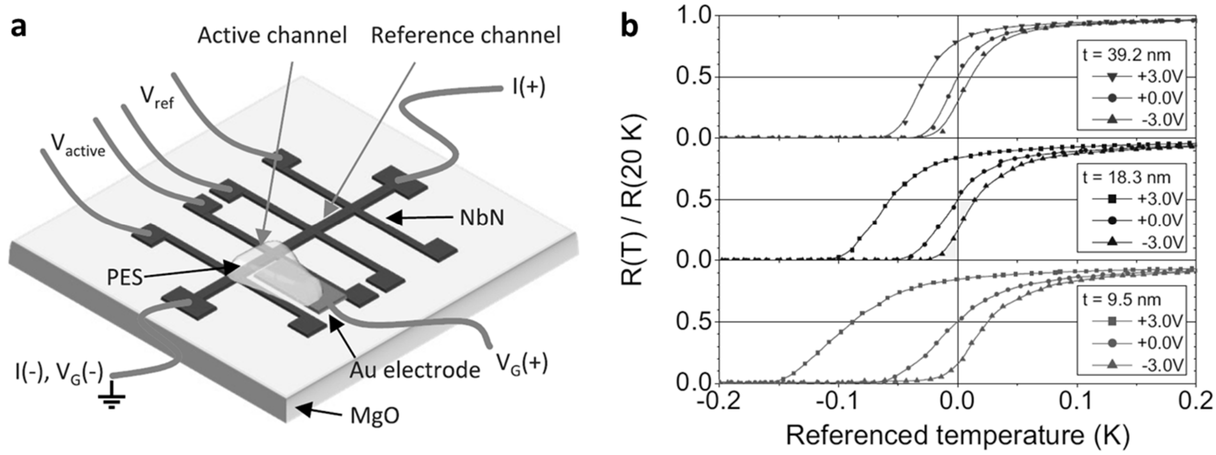

Figure 1: **a**, Sketch of a NbN-based EDL-FET in the dual channel configuration (active + reference). **b**, Normalized resistance of the active channel as a function of the temperature referenced to the midpoint of the ungated transition, for different applied gate voltages and film thicknesses. Adapted from Ref. 9.

We started our investigation by fabricating EDL-FETs on NbN thin films with a total thickness of ~ 40 nm, and surprisingly observed that no increase in T_c occurred upon surface electron accumulation. After developing a dual-channel architecture (Fig.1a) to allow for an accurate measurement of very small T_c shifts, upon electron accumulation we were able to observe only small *suppressions* ($\approx$0.3%) of T_c instead, whereas small *enhancements* of T_c were observed only upon electron depletion (Fig.1b, top panel) [8]. The observation of T_c suppressions was particularly puzzling, as it required the SC pairing to be suppressed in the whole thickness of the film to be detected, since a surface-bound suppression of T_c would be shunted by the underlying unperturbed bulk.

If the perturbation to the SC condensate extends to the entire bulk of the film, the T_c shifts for a given value of the induced carrier density will be strongly dependent on the film thickness. We thus employed ion milling to progressively thin down our NbN films from ~ 40 to ~ 10 nm and observed that indeed the magnitude of the T_c shifts increased upon decreasing the film thickness, as expected (Fig.1b, middle and lower panels) [9]. These findings confirmed that, despite the gate-induced electric field being confined in a thin layer at the surface by electrostatic screening, the perturbation to the superconducting state extends in a region much larger than a single unit cell. Indeed, the dependence of T_c on the induced carrier density and thickness could be reconciled with the strong-coupling BCS theory of superconductivity only if this thin surface layer is coupled to the underlying, unperturbed bulk via proximity effect [9].

By combining the strong-coupling BCS model for the proximity effect with ab-initio calculations of the doping-dependent density of states (DOS) in the system, we also determined that the thickness of this electronically-perturbed surface layer (i.e. the screening length of the electric field) strongly increases for large gate electric fields, reaching values of the order of 3 nm at the highest doping (Fig.2a) [9]. This unexpected feature could be partially accounted for by including non-linear corrections to the standard Thomas-Fermi screening model [9], and a more accurate treatment via state-of-the-art DFT calculations in the FET architecture reproduced the experimental results and linked this anomalous increase of the screening length to a distortion of the pristine charge density in the material upon the application of sufficiently large electric fields (Fig.2b) [10]. Additionally, we developed a comprehensive model combining DFT calculations of the electronic structure and Migdal-Eliashberg theory of the proximity effect to predict the electric-field-induced T_c shifts in any metallic superconductor of arbitrary thickness purely from first principles [11].

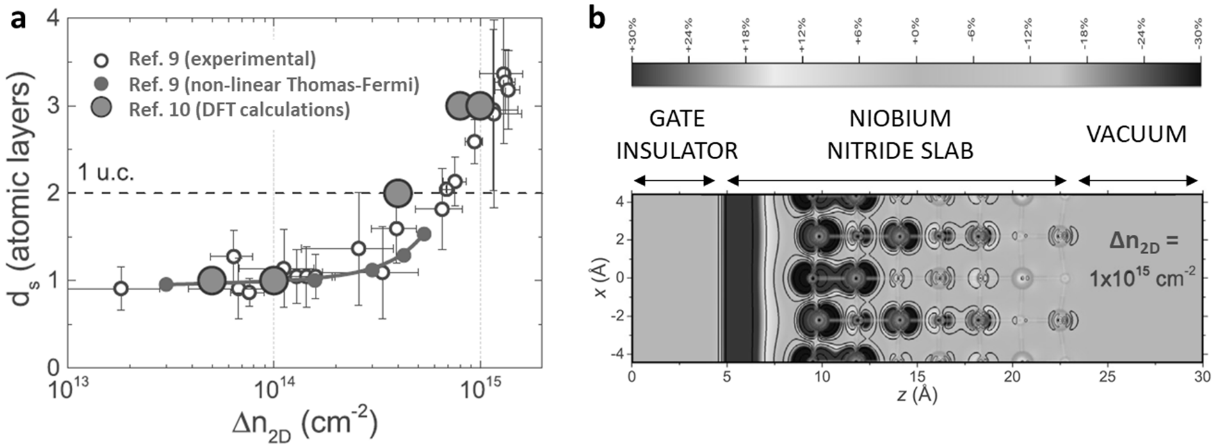

Figure 2: **a**, Number of atomic layers involved in the screening of the electric field, as a function of the induced charge density. **b**, Difference between the total charge density in doped and undoped NbN slab, sliced along the (x,z) plane. Grey-scale map is in linear scale between ±30% of the maximum density difference in the accumulation layer. Adapted from Ref. 10.

This coupling of the quasi-2D perturbation of the electronic structure to the long-range "bulk" modulation of the SC condensate stems from the intrinsically coherent nature of the latter, which cannot be perturbed over length scales smaller than its coherence length. It is therefore a general behavior in gated superconductors that could hinder the possibility to obtain large T_c shifts in films thicker than the screening length. Our current experimental focus is thus on exploring the tunability of ultrathin (< 5-nm-thick) NbN films to maximize the gate-induced T_c shift by minimizing the influence of the bulk. In doing so, we discovered that ultrathin NbN films are much less stable than their thicker counterparts, which prompted us to develop a novel technique of self-encapsulation in ultrathin niobium oxide to ensure the full reversibility of the gate modulation in these extremely sensitive devices. Our self-encapsulated ultrathin NbN films exhibited fully reversible modulations of both the normal-state resistance and the T_c, with gate-induced T_c shifts over three times larger than the maximum ones observed in unprotected, thicker devices, and for $\sim$10 times smaller induced carrier densities.

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