InAs/MoRe hybrid semiconductor/superconductor nanowire devices

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Implementing superconductors capable of proximity-inducing a large energy-gap in semiconductors in the presence of strong magnetic fields is a major goal towards applications of semiconductor/superconductor hybrid materials in future quantum information technologies. Here, we study the performance of devices consisting of InAs nanowires in electrical contact to molybdenum-rhenium (MoRe) superconducting alloys. The MoRe thin films exhibit transition temperatures \( T_C \) and critical fields exceeding 6 T. Normal/superconductor devices enabled tunnel spectroscopy of the corresponding induced superconductivity, which was maintained up to \( T_C \) and MoRe based Josephson devices exhibit supercurrents and multiple Andreev reflections. We determine an induced superconducting gap lower than expected from the transition temperature, and observe gap softening at finite magnetic field. These may be common features for hybrids based on large gap, type-II superconductors. The results encourage further development of MoRe-based hybrids.

The potential for building topologically protected quantum information processors from low-dimensional semiconductor-superconductor hybrid materials\(^{12}\) drives significant efforts across a range of fields including experimental and theoretical physics, device engineering, and material science.\(^{3}\) The main branch of experimental studies centers around hybrid semiconductor-superconductor devices employing a low band-gap, strong spin-orbit semiconductor such as InAs or InSb electrically connected to conventional superconductors such as aluminum\(^{4}\) or niobium.\(^{5}\) Aluminium is a type-I superconductor with a transition temperature of \( T_C \approx 1.5 \) K and low bulk critical magnetic field \( B_C \approx 10 \) mT. Although restricting to thin (\( \lesssim 10 \) nm) films and in-plane fields allows operating devices in high fields, obviating these restrictions motivates the development of hybrid materials with higher transition temperatures and bulk compatibility with metallic films.\(^{6–8}\) Type-II superconducting films of niobium and niobium based alloys (NbT, NbTIN) are well developed for metallic superconducting devices and have been incorporated in semi-super hybrid materials and devices.\(^{9–11}\) However, despite a bulk \( T_C \gtrsim 9 \) K, the induced superconducting gap in mesoscopic hybrids is often reduced to values comparable to those of Al.\(^{12}\) This is typically attributed to the formation of Nb oxides, which are superconducting with a lower \( T_C \approx 1.4 \) K, metallic, or magnetic\(^{12}\) and prompts efforts to develop semiconductor/superconductor hybrids where the favourable bulk superconducting properties are retained at the mesoscale.\(^{9–11}\) Since epitaxy between the highest single-element \( T_C \) superconductor and InAs has been demonstrated,\(^{5}\) focus must now turn to compound superconductors to further increase \( T_C \).

Here we investigate type-II molybdenum-rhenium (MoRe) alloys with high \( T_C \approx 10 \) K and \( B_C > 6 \) T\(^{13}\) as alternative materials for proximitizing InAs nanowires (NWs). The investigation of MoRe is further motivated by recent demonstrations of high quality MoRe superconducting resonators\(^{14}\) and bulk-like superconducting properties in very thin (4 nm) films, even with a relatively large oxygen content \( \approx 10 \)%\(^{15}\), suggesting tolerance to impurity incorporation exceeding that of Nb-based films. Finally, MoRe has been used as an electrode material in graphene,\(^{16–18}\) carbon nanotubes,\(^{21}\) and molecular\(^{22}\) superconducting devices, enabling operation in high magnetic field. However, a detailed spectroscopic study of the (sub-)gap properties of induced superconductivity from MoRe contacts has not been undertaken, and devices featuring semiconductors with large \( g \)-factors and strong spin-orbit coupling\(^{19,20}\) are lacking. We therefore fabricated MoRe/InAs-nanowire/TiAu (S-NW-N) tunneling spectroscopy devices and MoRe/InAs-nanowire/MoRe (S-NW-S) Josephson devices and demonstrate crucial features of hybrid devices, including an induced superconducting gap present up to \( T_C \), sub-gap states and multiple Andreev reflections, with high yield. Unexpectedly, we find a reduced induced superconducting gap compared to that expected from the measured \( T_C \) of the contacts and a softening of the superconducting gap at low fields compared to \( B_C \) of the contacts. We discuss possible reasons for these observations and future directions.

MoRe thin films were deposited by DC magnetron sputtering from a commercially available target of \( \text{Mo}_0.3\text{Re}_{0.7} \) using a power of 150 W under a 4 mTorr of Ar. The resulting deposition rate was of 11 nm/min and 250 nm thick films were deposited on doped silicon substrates capped with 200 nm of SiO\(_2\) and patterning was achieved by conventional electron beam lithography and lift-off using a \( \sim 350 \) nm thick poly methyl methacrylate resist. A 250 nm film thickness was chosen to ensure complete nanowire coverage, matching the typical thicknesses for Ti/Au contacts.\(^{20,24}\) Devices for bulk characterization of the MoRe films were in the form of 3 \( \mu \)m wide MoRe strips measured in a four-terminal configuration with a 20 \( \mu \)m separation of the inner voltage
probes (see sketch on Fig. 1a).

For hybrid devices, InAs NWs were grown by molecular beam epitaxy using Au catalyst particles following standard recipes. NWs have diameters $d_{NW} = 100 - 115$ nm and lengths $\sim 10 \mu$m. Individual NWs were transferred from the growth substrate to the device substrate using a manual micro-manipulator and located with respect to a predefined alignment grid by optical microscopy. S-NW-N and S-NW-S devices have electrode separations of $95 - 130$ nm and $140 - 365$ nm, respectively, and Ar$^+$ milling was performed before metal deposition with the aim of reducing native oxides and facilitating ohmic contacts. Figure 1 shows an SEM micrograph of a typical S-NW-N device. The electron density of the NW between the contacts was tuned by biasing the doped Si substrate at voltage $V_g$, and each electrode was split into two to allow a pseudo four-terminal measurement of the differential conductance $G = dI/dV$. As such, contributions from in-line resistances due to cryostat wiring/filtering are absent from measurements. Experiments were performed using standard lock-in techniques in a dilution refrigerator with a base temperature of $15$ mK.

To investigate the induced superconducting properties we focus first on the S-NW-N devices. Figure shows $G$ vs. $V_{sd}$ and $V_g$ close to pinchoff. In this regime, quantum dots (QD) form in the junction and when operated in Coulomb blockade, the QD acts as a tunnel barrier and the differential conductance, $G = dI/dV$, measures the density of states of the proximitized InAs. The $V_g$-dependent features observed at low bias are associated with Yu-Shiba-Rusinov (YSR) states arising from hybridisation between the QD levels and the superconducting contact. Gate-independent features at higher bias can be associated with the superconducting gap, and/or bound states with an energy close to that of the gap. The induced superconducting gap is therefore estimated to be $\Delta^* \sim 0.9 \text{ meV}$, although better certainty may be obtained in future measurements by utilising a device geometry where the density of states underneath the supercon-
A key motivation for investigating MoRe as an electrode material is the compatibility of its superconducting phase with magnetic fields. Returning to Figs. 2c, d – which show bias spectra with $B_{\perp} = 1$ T and $B_{\perp} = 2$ T, respectively – we observe that although the MoRe films are superconducting to $\sim 100$ K, the superconductivity-related features around zero bias are strongly suppressed (white arrows) compared to the zero field case in Fig. 1a. The field dependence is detailed in Fig. 3b, which show measurements of $G(V_{sd})$ vs. $B_{\perp}$ and $B_{\parallel}$, respectively, at a fixed $V_{g} = 1.8$ V within a CB diamond corresponding to even occupancy of the QD (red arrow in Fig. 4a). Here $B_{\parallel}$ is the in-plane field parallel to $B_{\perp}$.
the axis of the NW. In both cases, states detach from the gap edge and fill the gap at ~ 0.35 T and 0.45 T for $B_{\perp}$ and $B_{\parallel}$, respectively. This evolution is emphasized in Fig. 3, showing extracted $G(V_{sd})$ for different fields, and in Fig. 2a, which shows $G_S$ and $G_N$ as a function of $B$. A reduced gap remains visible and $G_N/G_S$ rapidly decreases for fields above 0.25 T. Similar observations have been reported for devices based on type-II superconductors Nb, NbTi, and NbTiN and associated to the formation of vortices in the electrodes, causing $\Delta$ to locally vanish thus softening the superconducting gap and/or shifting spectral weight from the coherence peaks to energies within the gap. We note, that for $B = 210 \text{ mT}$ the average vortex separation $\sqrt{4a/3B}$ reaches the NW diameter and thus $\Delta = 0$ vortex cores will be found proximal to the nanowire tunnel probe, softening the local density of states measurement.

Figure 3 show measurements similar to Fig. 2a,c,h except measured as a function of temperature and for $V_g = 2.38 \text{ V}$ (green arrow in Fig. 2a). The coherence peaks weaken with temperature and the in-gap conductance increases markedly from ~ 1 K and reaches $G_N$ at ~ 10 K (see Supporting Information Fig. S5) consistent with $T_C$ of the bulk MoRe film. Assuming $\Delta^*$ is temperature independent at $T \ll T_{\text{bulk}}$ the zero bias conductance was fitted to the conventional expression for the N-S tunnelling $\frac{G_S|_{V_{SD}=0}}{G_N} = G_N \sqrt{2\Delta^2/(k_B T)} \exp(-\Delta^*/k_B T)$. The solid red (dashed orange) line in Fig. 3g shows extrapolation of a fit performed up to 1.5 K (3 K), yielding $\Delta^* = 0.52 \text{ meV}$ (0.62 meV). The dependence on fitting range, relatively poor fit at low $T$ (see Supporting Information for data plotted on a linear scale), and low values of $\Delta^*$ compared to those obtained from bias spectroscopy, suggests this fit may be affected by additional contributions to the conductance in addition to thermal excitation above $\Delta^*$, likely mediated by a high density of sub-gap states due to the non-ideal MoRe/InAs interface as discussed in connection with Fig. 3.

Future experiments involving high-purity interfaces may solve this issue.

Transport in S-NW-S Josephson devices where both electrodes are made from MoRe is governed by different phenomena than in S-NW-N devices: Dissipation-less supercurrents carried by Andreev bound states dominate at zero bias, while multiple Andreev reflections dominate at finite bias. Figure 4a shows the differential resistance $dV/dI$ of S-NW-S device 4 as a function of $V_g$ and the current $I$ through the device in a regime far from pinch-off ($G_N \geq 2 e^2/h$). Here, $V$ is the measured voltage drop across the device, and measurements were performed in current-bias configuration with a 200 MΩ
bias resistor (see Supporting Information Figure S4). A zero-resistance state is observed at low currents with a switching current $I_{sw}$ (white line) which decreases upon reducing $V_g$ due to the depletion of the NW. In addition, $I_{sw}$ exhibits modulations with $V_g$ due to conductance fluctuations in the coherent InAs NW. In Fig. 4, the current was swept from negative to positive and the minimal switching/retrapping hysteresis indicates overdamped junction dynamics and/or Joule heating. A maximal $I_C = 23 \text{nA}$ was found at $V_g = 20 \text{ V}$ with a product $I_C R_N = 0.1 \text{mV} \ll \pi \Delta / (2e) = 2.2 \text{ mV}$. The critical currents of nanowire JJs have been widely investigated and similar to the present findings, $I_C R_N$ is commonly found to considerably underestimate $\Delta$. A complete description of the underlying cause(s) is lacking, however disorder and/or inhomogeneity of materials and interfaces are likely an important factor.

Figure 4b shows the $dV/dI$ vs. $B_L$ at $V_g = 20 \text{ V}$. The switching current $I_{sw}$ decreases monotonically as expected from diffusive transport in narrow, long nanowire junctions where flux pick-up in the junction is suppressed. For a diffusive junction the decay is expected to follow the relation $I_{sw}(B_L) = I_{sw}^0 \exp \left[ -\frac{B_L}{B_{C}} \right]$, with flux quantum $\phi_0 = h / 2e$ and $I_{sw}^0 = I_{sw}(B_L = 0)$. The dashed line in Fig. 4b shows a fit to the measured $I_{sw}$ with a fixed NW diameter $d_{NW} = 115 \text{ nm}$ determined from SEM images of Dev. 4 and taking the length of the NW junction, $L$, as a free parameter. The fitted $L \sim 240 \text{ nm}$ is larger than the contact separation $\sim 175 \text{ nm}$ as previously observed for devices with a substantial semiconductor/superconductor overlap. This supports that the reduction of the measured induced gap, $\Delta^*$, in the S-NW-N may be influenced by a decay from the electrode to the QD position as discussed above. The temperature dependence of $I_{sw}$ in Fig. 4c exhibits a decay typical for a diffusive junction and similar to the observed $I_{sw}$ suppression below $B_{C}^{bulk}$, complete suppression of $I_{sw}$ occurs for $T \approx 3 \text{ K} < T_C$ around the same temperature where $\Delta^*(T)$ vanishes – cf. Fig. 3f.

Finally, we show in Fig. 4e bias spectroscopy in a regime close to pinch off $V_g \lesssim 0 \text{ V}$ where the device is dominated by a quantum dot in Coulomb blockade analogous to Fig. 2. For $V_g$ and $V_{sd}$ inside Coulomb blockade, sequential transport is suppressed, and the current is mediated by co-tunneling. As typical for this situation, the conductance exhibits gate independent structure at finite bias $eV_{sd} = \pm 2\Delta^*/n, n = 1, 2, 3, \ldots$ due to the successive opening and closing of the allowed multiple Andreev reflection (MAR) processes. Since the MAR processes in Coulomb blockade regime occur across a randomly located quantum dot, the superconducting leads consist of the proximitized NW segments either side of the dot to thus analysis of the MAR allows an alternative method for determining $\Delta^*$. Figure 4e shows the extracted $G(V_{sd})$ averaged over the $V_g$-range indicated by a white arrow in Fig. 4b. A series of peaks are observed following the expected harmonic series with the indicated MAR order $n$. The inset shows the extracted peak positions as a function of $1/n$ and a linear fit gives $\Delta^* = 1.1 \text{ meV}$. This value is consistent with that obtained from N-NW-S tunnel spectroscopy given the observed sample to sample variation and close to the value obtained for MoRe/graphene devices. Continuous tunability between the ‘open’ and Coulomb blockade regimes was seen in all devices (Supporting Figs. S2,S3).

To summarize, we have investigated the sputtered alloy molybdenum-rhenium Mo$_{45.5}$Re$_{54.5}$ as a superconducting contact material to InAs nanowires. Both MoRe-NW-NW tunnel spectroscopy devices and MoRe-NW-MoRe Josephson devices were successfully fabricated and electrically characterized at cryogenic temperatures. The
MoRe electrode films have a transition temperature of 9–10 K corresponding to a superconducting gap $\Delta_{\text{bulk}} = 1.4$ meV, and the critical field exceeds 6 T. S-NW-N tunnel spectroscopy and analysis of the sub-gap structure in the conductance of S-NW-S devices shows an induced superconducting gap in the NW of $\sim 0.5–1.1$ meV. This is lower than $\Delta_{\text{bulk}}$ which we associate with disordered MoRe/NW interfaces or effects of a diffusive junction. Applying a magnetic field, we find that features associated with induced superconductivity are visible even for high perpendicular fields $B_{\perp} > 2$ T, however, the sub-gap conductance of the tunnel devices increases and the critical current of JJ devices decreases rapidly already within 500 mT. This finding is similar to previous studies of type-II superconductor/semiconductor hybrid devices. Although the nanowire exhibits a soft, ‘V’-shaped gap, we expect that the hardness could be improved by careful cleaning of the semiconductor interface or by in-situ deposition after nanowire growth. MoRe thus appears a promising candidate to extend semiconductor–superconductor hybrids towards higher $T_C$ operation, and potentially extended magnetic field ranges, if the role of vortices can be elucidated through, e.g., STM studies. In particular, we expect device performance to improve further if shadow epitaxy technique can be applied to the sputter deposited films. In the short term, our results suggest MoRe as an alternative to Nb or NbTiN, offering simple processing, with superconducting properties less sensitive to oxide formation, and larger residual resistance ratios for RF applications.

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COMPETING INTERESTS

The authors declare no competing interests.

SUPPORTING INFORMATION

The Supporting Information contains circuit diagrams, additional tunnel spectroscopy data, additional analysis of the data in Fig. 3g and the raw data of Fig. 4d. This material is available free of charge via the internet at https://doi.org/10.1021/acs.nanolett.2c02532.
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