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# Hard Superconducting Gap and Diffusion-Induced Superconductors in Ge–Si Nanowires

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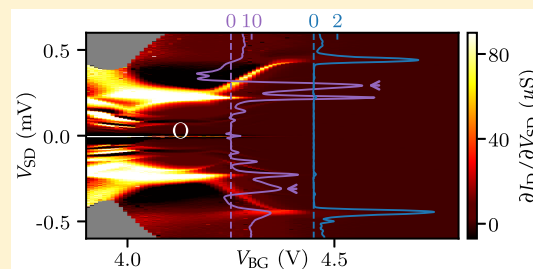
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## Supporting Information

**ABSTRACT:** We show a hard superconducting gap in a Ge–Si nanowire Josephson transistor up to in-plane magnetic fields of 250 mT, an important step toward creating and detecting Majorana zero modes in this system. A hard gap requires a highly homogeneous tunneling heterointerface between the superconducting contacts and the semiconducting nanowire. This is realized by annealing devices at 180 °C during which aluminum interdiffuses and replaces the germanium in a section of the nanowire. Next to Al, we find a superconductor with lower critical temperature ( $T_C = 0.9$  K) and a higher critical field ( $B_C = 0.9–1.2$  T). We can therefore selectively switch either superconductor to the normal state by tuning the temperature and the magnetic field and observe that the additional superconductor induces a proximity supercurrent in the semiconducting part of the nanowire even when the Al is in the normal state. In another device where the diffusion of Al rendered the nanowire completely metallic, a superconductor with a much higher critical temperature ( $T_C = 2.9$  K) and critical field ( $B_C = 3.4$  T) is found. The small size of these diffusion-induced superconductors inside nanowires may be of special interest for applications requiring high magnetic fields in arbitrary direction.

**KEYWORDS:** Superconductor–semiconductor hybrid device, topological superconductivity, Majorana quasiparticle, Ge–Si nanowire, Josephson junction, hard superconducting gap



The discovery that Majorana fermions offer a route toward an inherently topologically protected fault-tolerant quantum computer<sup>1–3</sup> marked the beginning of a quickly growing field of research to achieve their experimental realization. Majorana fermions require a topological superconducting material, which in practice can be realized by coupling a conventional *s*-wave superconductor to a one-dimensional nanowire with high spin–orbit coupling and *g*-factor.<sup>4–7</sup> Signatures of Majorana fermions are expected to arise as a conductance peak at zero bias and finite magnetic fields. The first reports showing these zero-bias conductance peaks in InAs and InSb nanowires<sup>8–14</sup> suffered from sizable subgap conductivity attributed to inhomogeneities in the nanowire–superconductor interface.<sup>15,16</sup> The resulting quasiparticle poisoning decoheres Majorana states since they will participate in braiding operations<sup>17–19</sup> and additionally obscure the Majorana signatures at zero energy. Strong efforts have been made to improve these interfaces, that is, induce a hard gap, using epitaxially grown Al<sup>20,21</sup> or specialized surface treatments methods,<sup>22,23</sup> resulting in much better resolved Majorana signatures.<sup>18,24–26</sup>

In contrast to the group III–V materials used in most previous work, we use Ge–Si core–shell nanowires consisting

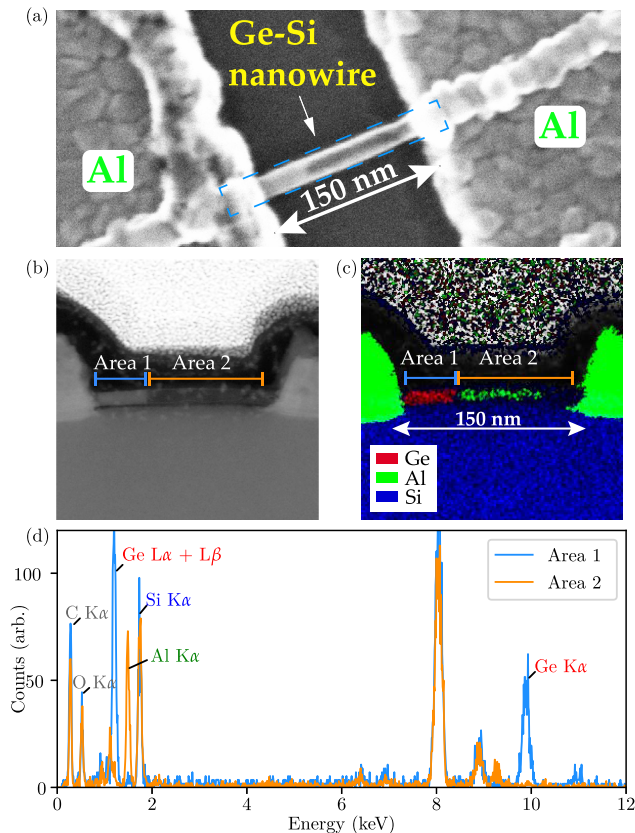
of a monocrystalline Ge  $\langle 110 \rangle$  core with a diameter of  $\sim 15$  nm and a Si shell thickness of 2.5 nm covered by a native SiO<sub>2</sub>. Coherent strain in the defect-free crystal structure results in high hole-mobilities.<sup>27</sup> The electronic properties of the one-dimensional hole gas localized in the Ge core<sup>28,29</sup> make them a candidate for observing Majorana fermions,<sup>30,31</sup> although their interaction with a superconductor is still relatively unexplored.<sup>32–36</sup> These wires are predicted to have a strong first-order Rashba type spin–orbit coupling<sup>37</sup> which, together with the *g*-factor,<sup>38,39</sup> is tunable by electric fields. Our devices consist of a nanowire channel with superconducting Al source and drain placed on an oxidized Si substrate (for more detailed information about the fabrication process see Supporting Information Section SI). We focus on two devices where an essential thermal annealing process results in interdiffusion between Al in the contacts and Ge in the nanowire channel. Device A is an electric-field tunable Josephson junction<sup>34,36</sup> as shown in Figure 1a, whereas in device B the whole

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semiconducting nanowire channel has been metalized and we suspect Al has largely replaced the semiconductor.



**Figure 1.** Al–Ge interdiffusion in device A. (a) Top view SEM image of the device showing a Ge–Si nanowire between two Al contacts. In the right part of the nanowire, a slightly darker contrast is observed (see Figure S2 for SEM images showing this effect in several devices). The blue dashed line shows the approximate location of the TEM lamella. (b) Cross-sectional HAADF-STEM image of the same device. The same contrast difference as in (a) is observed. (c) HAADF/STEM image with combined EDX data for elements Ge, Al, and Si (see Figure S1 for separate images). (d) EDX spectrum for Area 1 and Area 2 as defined in (c).

The electric field dependence of Device A has already been extensively studied in ref 34 where the main result was the observation of two distinct regimes: a highly transparent regime with a near ideal  $I_C R_N$  product in accumulation, and a tunneling regime with few-hole occupancy where supercurrent only appears at the charge degeneracy points. In this work, we extend on this by investigating the magnetic field dependence of the transport properties in both regimes.

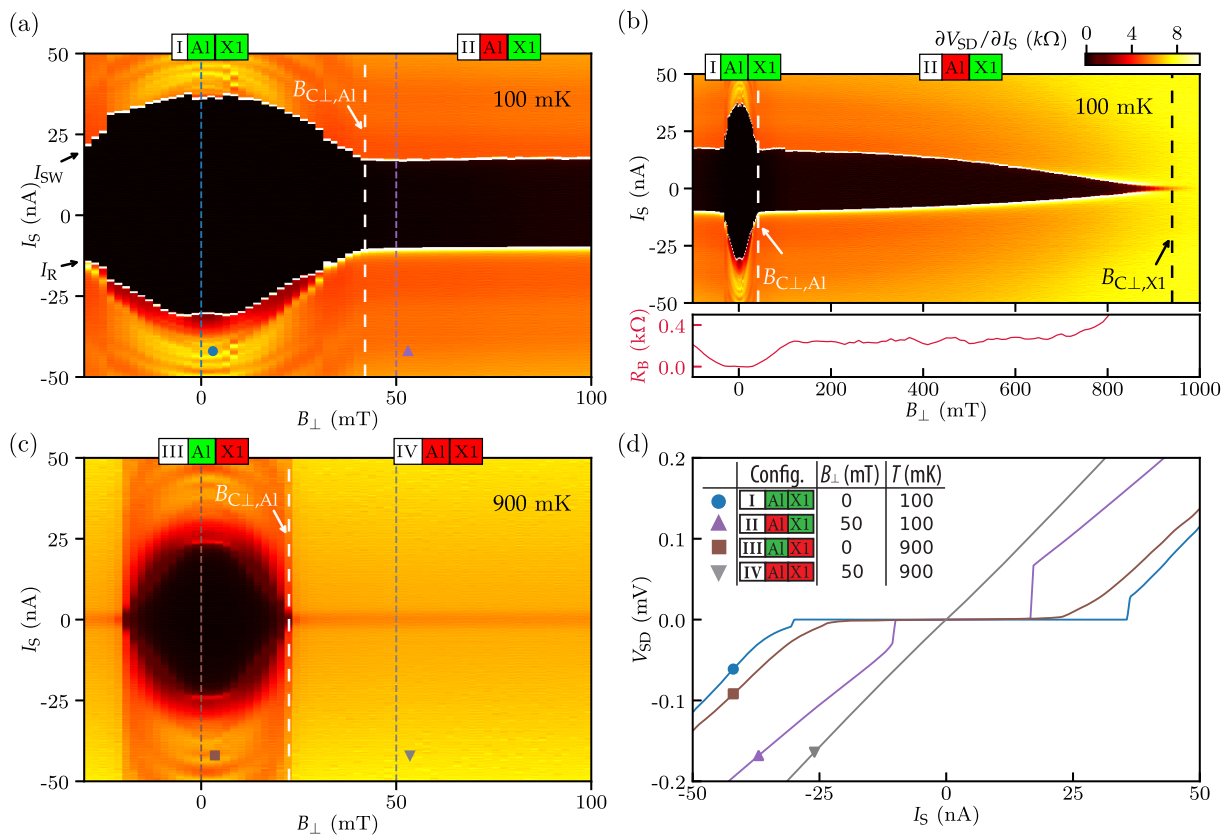
To gain insight into the microscopic properties of the superconductor–semiconductor interfaces, we start by investigating Device A using high-angle annular dark-field–scanning transmission electron microscopy (HAADF-STEM) in combination with energy-dispersive X-ray spectroscopy (EDX). We find strong indications that the additional superconductor, as well as the highly homogeneous superconductor–nanowire interface arises during the thermal annealing process where Al interdiffuses with the material in the semiconducting nanowire. In the second part, we map the switching current  $I_{SW}$  as a function of critical field  $B_C$  and critical temperature  $T_C$  of device A and B, which clearly shows an additional super-

conducting phase in both devices. In the final part, we investigate the hardness of the superconducting gap in the semiconducting nanowire of device A, by means of electronic transport measurements near depletion,<sup>20,23</sup> and observe that the conductance in the gap is suppressed by a factor  $\sim 1000$ .

**Al–Ge Interdiffusion.** To investigate the effects of the annealing on the stoichiometric composition of the nanowire channel, a TEM lamella was made along the nanowire axes of device A as indicated in Figure 1a. We first apply a stack of protective SiO<sub>2</sub> and Pt layers and subsequently create the TEM lamella using a standard focused ion beam lift-out protocol. This allows us to perform an analysis on the cross-section of the device, as can be seen in Figure 1b. In both panels a and b in Figure 1, a smaller region (Area 1) with higher contrast on the left and a bigger region with lower contrast on the right (Area 2) can be observed. Figure 1c shows the resulting EDX signals in these regions for the elements Ge, Si, and Al, and we observe the following clear distinction: in Area 1 we observe a strong Ge signal whereas in Area 2 the signal is dominated by Al.

In Figure 1d, we show the integrated EDX spectra for both areas. When comparing the two areas, we observe that in Area 2 the Ge L $\alpha$ , Ge L $\beta$ , and Ge K $\alpha$  signals fall below the detection limit. As is the convention in EDX analysis, L and K denote the orbital to which an electron decays in a picture where K, L, and M are the outer atomic orbitals, whereas  $\alpha$  and  $\beta$  indicate whether it decays from the first or second higher orbital. The Al K $\alpha$  signal shows the opposite behavior, implying that Ge has been replaced by Al in Area 2. The counts for elements O, C, and Si remain equal in both areas (see also Figure S1). As we will discuss in the following section, the superconductor in Area 2 has profoundly different properties from the Al contacts and we therefore refer to it as X1. Interdiffusion has also taken place below the left contact without reaching the channel, although this is not evident from the TEM data. Instead, we conclude this from transport data in the next section (Figure 2 and Figure S3). As a side-note, we cannot observe the effects of the interdiffusion process on the Si shell, because the Si signal is dominated by the SiO<sub>2</sub> that covers the substrate.

An in-depth study on the thermally induced interdiffusion process between Al and pure Ge  $\langle 111 \rangle$  nanowires, a highly similar system to ours, has been performed in refs 40 and 41. Here, in situ monitoring of the metal front inside the nanowires at various temperatures reveals that the velocity of propagation as a function of the length of the metalized nanowire segment is volume-diffusion limited and possibly surface-diffusion limited with the Al forming a monocrystalline face-centered cubic crystal inside the nanowire. The metal front forms an atomically sharp interface and no intermetallic phase is found in the metalized nanowire segment, that is, the Ge is transported out of the wire into the Al contacts. These observations are explained by a 15 orders of magnitude lower diffusion constant for Al in Ge than for Ge in Al.<sup>42,43</sup> Furthermore, the initial start of the diffusion reaction is governed by the respective activation energies (121.3 kJ/mol for Ge in Al, 332.8 kJ/mol Al in Ge<sup>42,43</sup>) and may depend on the specific atomic arrangement of the initial nanowire–Al interface, explaining the variation in the starting time of the diffusion reaction, even for two separate contacts on the same wire. These findings largely correspond to our observations on Ge–Si core–shell nanowires and give an explanation for the asymmetry in our contacts (see Figure S2 for SEM images of



**Figure 2.** Device A: Josephson junction with two superconductors. (a) Differential resistance  $\partial V_{SD}/\partial I_S$  versus  $I_S$  and  $B_{\perp}$  taken at  $T = 100$  mK. Black region corresponds to superconductivity. The white dashed line indicates  $B_{C,L,Al}$ . Arrows indicate  $I_{SW}$  and  $I_R$ . (b) Top panel: Same as (a) for a larger range of  $B_{\perp}$ . The vertical black dashed line indicates  $B_{C,L,X1}$ . Bottom panel: Horizontal cross-section showing  $\partial V_{SD}/\partial I_S$  versus  $B_{\perp}$  taken at  $I_S = 0$ . The color scale also applies to (a,b). (c) Same as (a) taken at  $T = 900$  mK. (d) Combinations of Al and X1 in the superconducting (green boxes)/normal (red boxes) state are numbered as configurations I–IV. Linecuts showing  $V_{SD}$  versus  $I_S$  taken for each configuration at the corresponding symbols in (a,c). Inset: table summarizing the configurations and values of  $B_{\perp}$  and  $T$  for the respective linecuts. In all figures,  $V_{BG} = -4.7$  V and  $I_S$  is swept from negative to positive bias.

partly and fully metalized nanowires), as well as the variation in device properties.

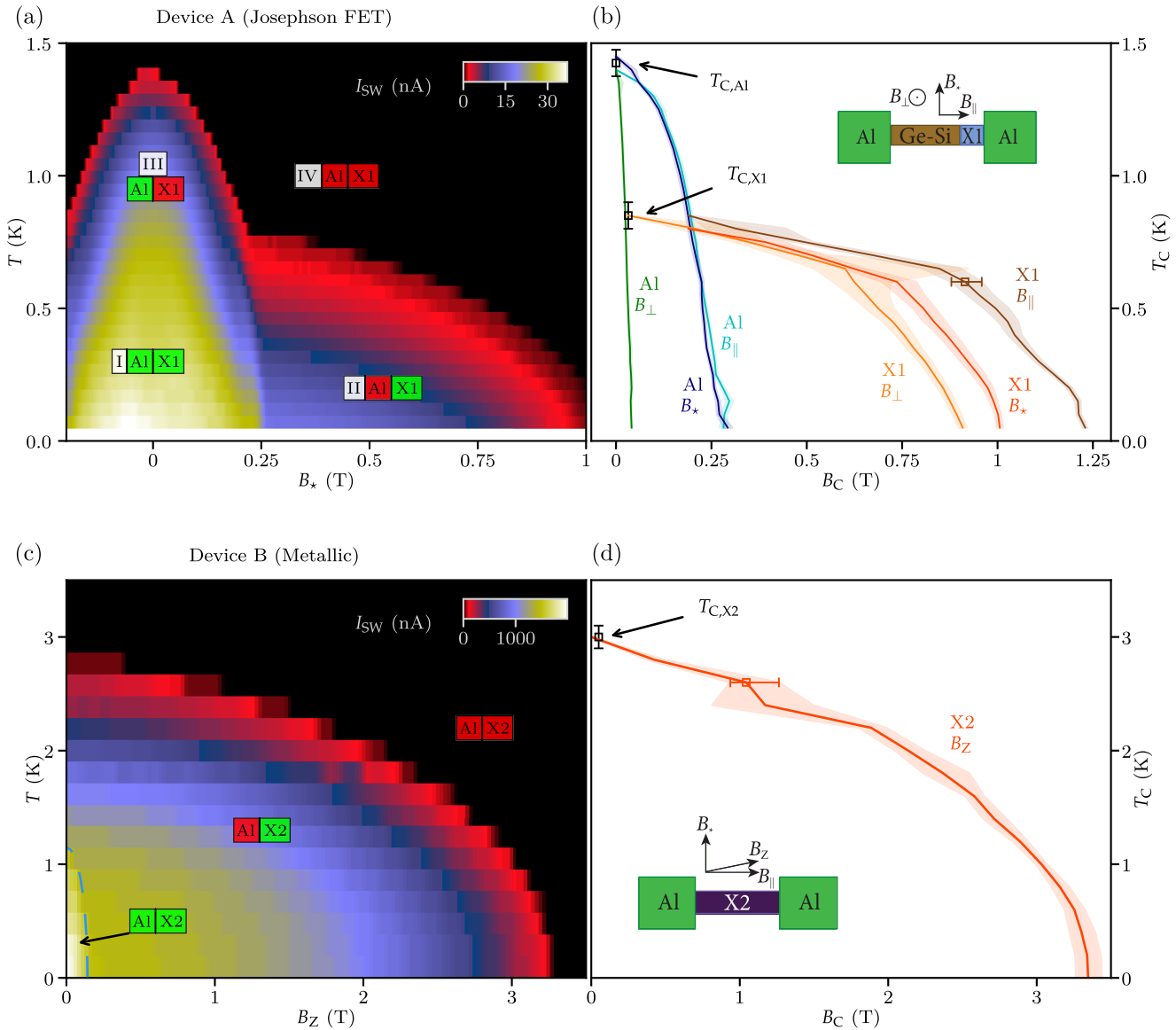
**Two Superconductors in a Nanowire Josephson Junction.** In Figure 2a, we show a magneto-spectroscopy of device A, the Josephson junction; we plot the differential resistance  $\partial V_{SD}/\partial I_S$  versus the sourced current  $I_S$  and the out-of-plane magnetic field  $B_{\perp}$  (see illustration in Figure 3b) while sweeping  $I_S$  from negative to positive current. The backgate  $V_{BG}$  is fixed at  $-4.7$  V where multiple subbands contribute to transport and the junction is highly transparent.<sup>34</sup> The superconducting region (black) is bounded by  $I_R < I_S < I_{SW}$  with  $I_R$  the retrapping current at negative bias and  $I_{SW}$  the switching current at positive bias. Upon increasing  $B_{\perp}$  from 0,  $I_{SW}$  decreases gradually until aluminum becomes normal at the critical out-of-plane field  $B_{C,L,Al} \approx 40$  mT after which a finite  $I_{SW}$  remains. For all  $B_{\perp}$ ,  $I_{SW} > I_R$  indicating that our junction is hysteretic for this particular value of  $V_{BG}$  due to the junction being underdamped<sup>34</sup> while additional heating-induced hysteresis can not be excluded<sup>44</sup> (see Figure S3a for a gate-dependence of  $I_{SW}$  and  $I_R$ ).

When increasing  $B_{\perp}$  further in Figure 3b,  $I_{SW}$  slowly decreases and finally disappears. The proximity-induced supercurrent above  $|B_{C,L,Al}|$  implies the presence of a second superconducting material, X1, in or near the nanowire channel with a critical field  $B_{C,L,X1} \approx 950$  mT. To confirm that our Al contacts are normal for  $B_{\perp} > |B_{C,L,Al}|$ , we consider the

background resistance  $R_B$  in the superconducting region as a function of  $B_{\perp}$  in the bottom panel of Figure 2b.  $R_B = 0$  for  $B_{\perp} < |B_{C,L,Al}|$ , whereas for  $B_{\perp} > |B_{C,L,Al}|$  the background resistance gradually increases to  $R_B \approx 0.25$  k $\Omega$  attributed to a normal series resistance of the Al contacts. Additionally, the out-of-plane critical field of a separately measured Al lead matches  $B_{C,L,Al}$  (see Figure S4).

In Figure 2c, we show a magneto-spectroscopy at 900 mK and observe that X1 is quenched for all  $B_{\perp}$ , while Al still induces a supercurrent for  $B_{\perp} < |25|$  mT. This shows that X1 has a lower  $T_C$  and a higher  $B_C$  than the Al contacts. Because X1 has a higher  $B_C$  and a lower  $T_C$  than Al, we can selectively switch either superconductor to the normal state, resulting in four possible device configurations I–IV as illustrated in Figure 2 and summarized in the inset in Figure 2d (a precise set of conditions for each configuration can be found in Table S1). Figure 2d shows plots of  $V_{SD}$  versus  $I_S$  in all four configurations, clearly showing a supercurrent in configuration II where Al is normal and only X1 is superconducting. Because we observe a gate-tunable Josephson current even in configuration II, we conclude X1 is present on both sides of the Ge–Si segment (see Figure S3 for differential resistance maps versus backgate in all four configurations).

**Junction  $I_{SW}$  versus  $B$  and  $T$ .** For the observed superconductors and their specific geometries, the critical field and critical temperature are interdependent variables and



**Figure 3.**  $I_{SW}$ ,  $T_C$ , and  $B_C$  of a Josephson FET (device A) and a metallized nanowire device (device B): (a)  $I_{SW}$  versus  $T$  and  $B_*$  for the Josephson FET (device A). The green (red) boxes indicate whether the material is superconducting (normal) and show the configurations I–IV as defined in the main text. (b)  $T_C$  versus  $B_C$  for Al and X1 for three main field axis  $B_{\perp}$ ,  $B_{\parallel}$ , and  $B_*$  as illustrated by the inset. Curves are extracted from plots such as (a) (see main text). (c)  $I_{SW}$  versus  $T$  and  $B_Z$  for the completely metallized nanowire (device B) consisting of alloy X2. The green (red) boxes indicate three possible configurations. For the configuration where Al is superconducting (for  $B_Z < 300$  mT and  $T < 1$  K) an enhancement of  $I_{SW}$  can be observed as denoted by the blue dotted line. (d)  $T_C$  versus  $B_C$  for X2 extracted from (c). Inset shows the in-plane  $B_Z$  field direction which is rotated  $\sim 10^\circ$  with respect to the nanowire.  $B_Z$  corresponds to the z-axis of the vector magnet, the only axis capable of fields  $> 1$  T. In both (b,d), the vertical error bar represents an uncertainty in  $T_C$  of  $\sim 3\%$  and shaded areas are standard deviations in  $B_C$  from fits.

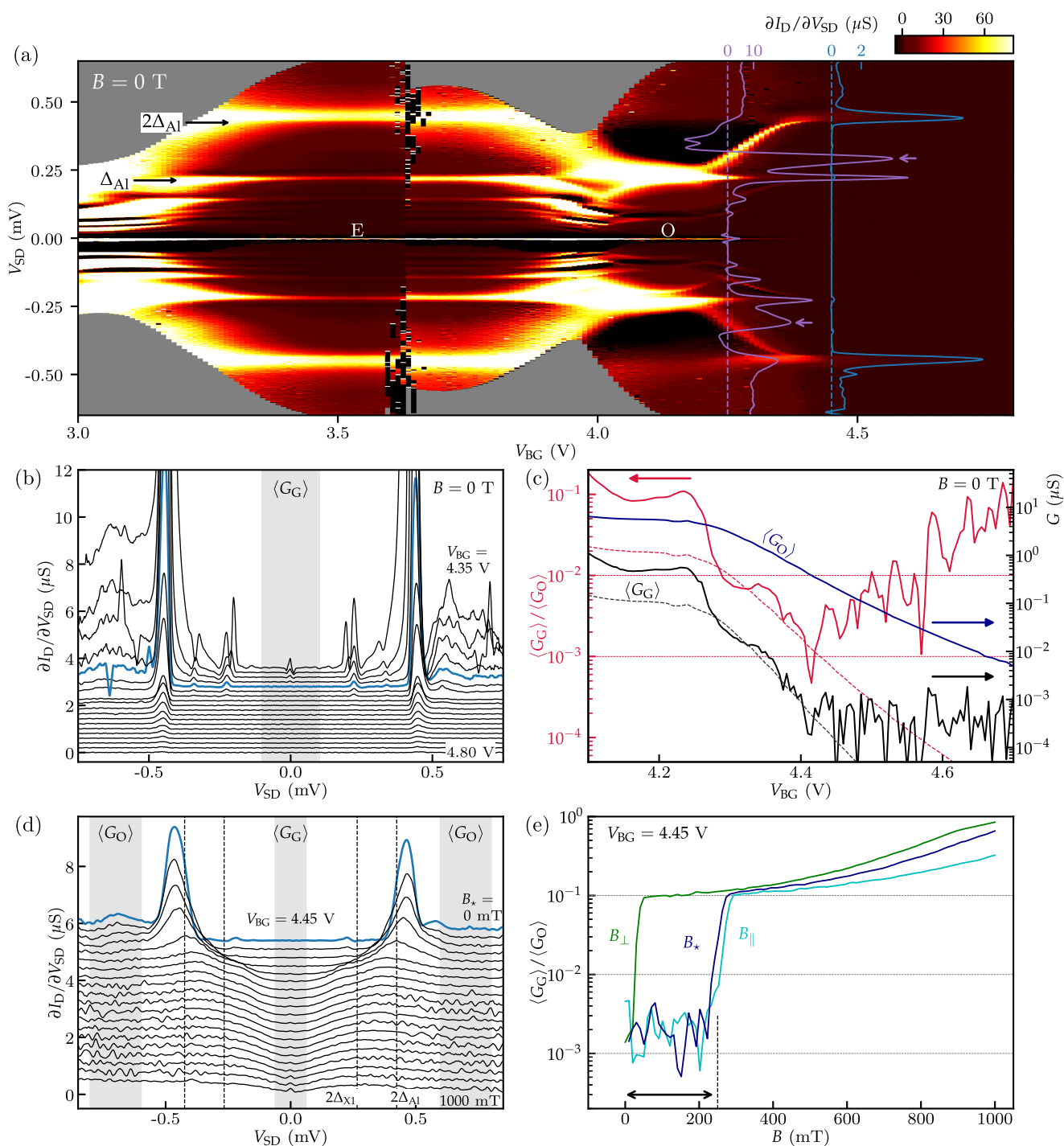
**Table 1.** Maximum values for  $T_C$ ,  $B_C$  of Al, X1, and X2 As Determined in Figure 3<sup>a</sup>

	$T_C$ (K)	$\Delta$ ( $\mu$ V)	$B_{C*}$ (mT)	$B_{C\perp}$ (mT)	$B_{C\parallel}$ (mT)
Al	$1.4 \pm 0.05$	$212 \pm 6$	$293 \pm 10$	$41 \pm 2$	$282 \pm 10$
X1	$0.9 \pm 0.05$	$133 \pm 8$	$1230 \pm 10$	$909 \pm 11$	$1010 \pm 20$
	$T_C$ (K)	$\Delta$ ( $\mu$ V)	$B_{Cz}$ (T)		
X2	$2.9 \pm 0.1$	$441 \pm 14$	$3.4 \pm 0.1$		

<sup>a</sup>We take  $T_{C,Al}(B_C = 0)$ ,  $T_{C,X1}(B_{\perp} = 50$  mT), and  $B_C(T \approx 0)$  to obtain their respective maximum values. The BCS superconducting gap is determined as  $\Delta = 1.764k_B T_C$ .

may have a nontrivial relation; the boundaries of the configurations I–IV in terms of  $B_C$  and  $T_C$  cannot directly be deduced from the data in Figure 2. We therefore collect  $I_{SW}$  versus  $B$  from magneto-spectroscopies for a large number of

temperatures and the three main magnetic field axes  $B_*$ ,  $B_{\perp}$ , and  $B_{\parallel}$  which are illustrated by the inset in Figure 3b. For the in-plane field perpendicular to the nanowire,  $I_{SW}$  has two clearly distinct overlapping shapes as a function of  $T$  and  $B_*$  in



**Figure 4.** Hard superconducting gap in a Ge–Si nanowire Josephson FET (Device A). (a) Differential conductance  $\partial I_D/\partial V_{SD}$  versus  $V_{SD}$  and  $V_{BG}$ . Odd (O) and even (E) hole occupation are denoted. The first two MAR orders are indicated at  $V_{SD} = 2\Delta_{Al}$  and  $\Delta_{Al}$ . (b) Vertical linecuts from (a) showing  $\partial I_D/\partial V_{SD}$  versus  $V_{SD}$  at 50 mV intervals in  $V_{BG}$ , curves are offset by 0.2  $\mu\text{S}$ . (c) Averaged in-gap conductance  $\langle G_G \rangle$  (black) and outside-gap conductance  $\langle G_O \rangle$  (blue) and the ratio  $\langle G_G \rangle/\langle G_O \rangle$  (red) versus  $V_{BG}$ . Dashed curves show theoretical minimal values and are the result of plotting eq 1. For every  $V_{BG}$ ,  $\langle G_G \rangle$  and  $\langle G_O \rangle$  are averaged over a range of  $V_{SD}$  as indicated by the gray area in (b) and the gray dashed lines in Figure S7, respectively. (d)  $\partial I_D/\partial V_{SD}$  versus  $V_{SD}$  for  $B_*$  from 0 to 1000 mT at 50 mT intervals. Curves are offset by 0.3  $\mu\text{S}$ . Dashed lines show the expected position of the quasiparticle peak for  $2\Delta_{Al}$  ( $2\Delta_{X1}$ ) at  $B = 0$ . (e) Ratio  $\langle G_G \rangle/\langle G_O \rangle$  for the three main field axes  $B_\perp$ ,  $B_\parallel$ , and  $B_*$  at  $V_{BG} = 4.45$  V (blue line in (a–c)). Ranges in  $V_{SD}$  where  $\langle G_G \rangle$  and  $\langle G_O \rangle$  are extracted are shown as gray areas in (d).

Figure 3a. The “peak” extending to  $T \approx 1400$  mK at  $B = 0$  with a width of  $|B_*| \approx 250$  mT at  $T = 50$  mK is attributed to the superconducting state of Al, whereas the second shape (the “tail”), extending up to  $\sim 1000$  mT at  $T = 50$  mK, corresponds to the superconducting phase of X1. We can thus map the four configurations in the color plot on the  $T$  versus  $B_*$  axes.

We now extract both the  $T_C$ – $B_{C*Al}$  and  $T_C$ – $B_{C*X1}$  curves from Figure 3a (see Supporting Information Section SII), that is, the critical temperature–critical field relation for Al and X1, and plot them in Figure 3b. We perform the same procedure for field directions  $B_\perp$  and  $B_\parallel$  (see Figure S5 for  $I_{SW}$  versus  $T$  and  $B_\parallel$  and  $B_\perp$ ).

In Table 1, we summarize the maximum  $T_C$ , the resulting superconducting gap  $\Delta$ , and  $B_C$  in the three field directions for Al and X1. Comparing  $B_{C\perp,Al} = 41$  mT with  $B_{C\star,Al} = 293$  mT and  $B_{C\parallel,Al} = 282$  mT we notice a factor  $\sim 7$  difference. This strong anisotropy for the out-of-plane field direction is clearly present in the  $T_{C,Al}-B_{C,Al}$  curves in Figure 3b and is expected for the large aspect ratio of the 50 nm thick Al contacts.

The  $T_{C,X1}-B_{C,X1}$  curves show a less prominent magnetic field anisotropy from which we can roughly deduce the shape of X1 by assuming that the normal surface of the material is inversely proportional to the critical field, that is, a larger superconducting normal-surface requires expelling more flux.<sup>45</sup> Using the respective ratios of  $B_{C\star,X1}$ ,  $B_{C\perp,X1}$ , and  $B_{C\parallel,X1}$ , we observe that X1 is slightly elongated along the nanowire axis, reaffirming the hypothesis that X1 resides in the nanowire channel.

We now switch to the completely metalized device B where we believe Al has diffused completely through the channel, effectively making the nanowire a metallic superconductor. Figure 3c shows  $I_{SW}$  versus  $T$  and  $B_Z$  where the corresponding  $T_{C,X2}-B_{C,X2}$  relation in Figure 3d is obtained by the previously mentioned polynomial fitting method. We see a critical temperature  $T_{C,X2} = 2.9$  K at  $B = 0$  and critical field  $B_{C,X2} = 3.4$  T at  $T = 50$  mK, both much higher than for X1 and the Al contacts. The switching current  $I_{SW} = 1.5$   $\mu$ A is 2 orders of magnitude higher compared to device A.

When comparing  $T_{C,X2} = 2.9$  K and  $B_{C,X2} = 3.4$  T with thin Al aluminum films,<sup>46</sup> we observe X2 has equivalent properties of an  $\sim 3$  nm thick film (in parallel field) and we could conclude that X2 is simply a very small cylinder of aluminum inside the nanowire channel. However, for X1 with  $T_{C,X1} = 0.9$  K and  $B_{C,X1} \approx 1$  T an equivalent film thickness cannot be defined. Even though no intermetallic phases were found for annealed pure Ge nanowires in refs 40 and 41, a possible origin of X1 is the formation of a Al-Si/Ge alloy in our core-shell nanowires, albeit with a ratio of semiconductor to Al below that of our EDX detection limit. In literature, certain stoichiometric compositions indeed result in a lower  $T_C$  than for pure Al,<sup>47,48</sup> and in fact one can get alloys with a  $T_C$  ranging from 0.5 K up to 11 K by various methods.<sup>49-53</sup> The exact composition of both X1 and X2 in our Ge-Si core-shell system therefore remains partly speculative and would require a more in-depth study like ref 41.

To sum up, we observe X1 with  $T_{C,X1} = 0.9$  K in a Josephson junction and X2 with  $T_{C,X2} = 2.9$  K in a metallic device, showing that diffusion of Al into Ge-Si nanowires can give rise to different superconductors with a  $T_C$  lower and much higher than that of the Al contacts, both appear as a second superconductor in transport measurements.

**Tunneling Regime of the Josephson FET.** We now focus on device A and tune  $V_{BG}$  to a regime where the nanowire is near depletion. Figure 4a shows the differential conductance  $\partial I_D/\partial V_{SD}$  versus the source-drain voltage  $V_{SD}$  and the backgate voltage  $V_{BG}$ . We notice a zero-bias conductance peak as the result of a finite Josephson current and a prominent multiple Andreev reflection (MAR) pattern showing as horizontal lines of increased conductance for  $V_{BG} = 3-4$  V. The reduced barrier transparency near depletion confines charges in the nanowire channel, and allows us to see odd and even charge occupation in a quantum dot in the wire<sup>34</sup> supported by a Kondo peak on the odd transitions<sup>34,54</sup> (see Supporting Information, Figure S6). Above  $V_{BG} = 4.4$  V, the MAR and zero-bias peak disappear, while the onset of

quasiparticle transport is visible at the superconducting gap at  $V_{SD} = \pm 2\Delta_{Al}$ . This trend is also present in the  $\partial I_D/\partial V_{SD}$  linecuts for  $V_{BG}$  between 4.35 and 4.80 V in Figure 4b.

In Figure 4a, between  $V_{BG} = 4.2$  V and  $V_{BG} = 4.4$  V we observe a conductance peak in both bias directions smoothly moving from  $|V_{SD}| = \Delta_{Al}$  to  $|V_{SD}| = 2\Delta_{Al}$ , when going from the odd to the even occupancy, which we attribute to an Andreev bound state (ABS). Additional evidence for an ABS presents itself in the form of a region of negative differential conductance in the odd occupancy between  $V_{SD} = \Delta_{Al}$  and  $V_{SD} = 2\Delta_{Al}$ ,<sup>55,56</sup> as highlighted by the purple linecut at  $V_{BG} = 4.25$  V in Figure 3a. Tunnel spectroscopy on an ABS requires asymmetric opaque tunnel barriers where the most opaque barrier probes the ABS.<sup>54</sup> A barrier asymmetry in our devices can indeed be expected, because the final interface properties are determined by microscopic details on the Al-nanowire interface during annealing. For lower  $V_{BG}$ , our barriers quickly become highly transparent<sup>34</sup> and we therefore only observe the ABS signature near depletion.

In contrast to the bias-symmetric MAR features, the asymmetric barriers show up in the intensity of the ABS signatures (see the arrows on the purple linecut in Figure 4a). Depending on the bias direction, there are two different rate-determining tunnel sequences: (1) tunneling through an opaque barrier onto a single ABS or (2) tunneling from an ABS through an opaque barrier into the Fermi sea. Sequence (2) has a much higher tunnel probability than (1), which results in the observed asymmetry in conductance.

**Hard Superconducting Gap.** A measure for the amount of quasiparticle states inside the gap is the in-gap suppression of conductance also termed as the hardness of the gap. We therefore investigate the ratio  $\langle G_G \rangle / \langle G_O \rangle$  where  $\langle G_G \rangle$  ( $\langle G_O \rangle$ ) is a conductance value inside (outside) the gap averaged over a range of  $V_{SD}$  as shown in Figure 4b.  $\langle G_O \rangle$  is determined from a similar measurement at higher bias (see Figure S7), sufficiently far away from  $2\Delta_{Al}$ . Figure 4c shows  $\langle G_G \rangle$ ,  $\langle G_O \rangle$ , and the ratio  $\langle G_G \rangle / \langle G_O \rangle$  versus  $V_{BG}$  and we find the conductance is suppressed by a factor of  $\sim 1000$  for  $V_{BG} \approx 4.4$  V which is an order of magnitude higher than previously reported in this system in the same superconductor-normal-superconductor (SNS) configuration.<sup>35</sup>

A SNS junction can naively be viewed as two superconductor-normal (SN) junctions in series and the theoretical dependence of  $G_G$  on  $G_O$  can therefore be approximated as<sup>57</sup>

$$G_{G,SNS} \approx \frac{G_{G,SN}}{2} = \frac{2e^2}{h} \frac{G_O^2}{\left(\frac{4e^2}{h} - G_O\right)^2} \quad (1)$$

and it follows that the equivalent conductance suppression of an SN device is a factor of two lower than for an SNS device. We use the averaged  $\langle G_O \rangle$  as  $G_O$  and obtain the theoretical minimal in-gap conductance  $G_{G,SNS}$ , as well as the corresponding ratio  $G_{G,SNS}/G_O$ , shown as dashed lines in Figure 4c. We find that above  $V_{BG} = 4.25$  V, the measured  $\langle G_G \rangle$  and  $\langle G_G \rangle / \langle G_O \rangle$  closely follow the theoretical curves until the noise limit of our equipment is reached for  $V_{BG} > 4.4$  V. This suggests that  $\langle G_G \rangle$  is not dominated by quasiparticle poisoning and that our superconductor-semiconductor interfaces do not facilitate inelastic scattering and have low disorder.<sup>15</sup> We note that for these values of  $V_{BG}$ , the Ge-Si island is not fully depleted ( $\langle G_O \rangle$  still decreases as a function of  $V_{BG}$  and can be fully suppressed) and transport takes place through a tunnel-

broadened quantum dot level (also see ref 34). However, the obtained theoretical minimal in-gap conductance should be considered an approximation because we do not take into account any difference in interface transparency between the two contacts.

When measured in a SNS configuration, the ratio  $\langle G_G \rangle / \langle G_O \rangle$  gives an upper limit and could in reality be lower because it can be increased due to several other reasons than quasiparticle poisoning. (1) For higher  $V_{BG}$ ,  $\langle G_G \rangle$  is limited by the noise floor of our measurement setup and does not further decrease. The decrease of  $\langle G_O \rangle$  now lowers the observed current suppression  $\langle G_G \rangle / \langle G_O \rangle$ . (2) For lower  $V_{BG}$ , MAR and the zero-bias peak, both characteristic for Josephson junctions, appear as conductance peaks inside the gap which leads to a decreased  $\langle G_G \rangle / \langle G_O \rangle$ . (3) The quantum dot in the junction may lead to Fabry–Perot resonances and Kondo-enhanced tunnelling around zero bias (see Figure S6). SN devices will not exhibit these effects and may therefore result in a lower ratio  $\langle G_G \rangle / \langle G_O \rangle$  and give a better approximation of the quasiparticle density in the gap. Because of this, we cannot directly compare the current suppression in our device with other work probing the superconducting gap using a single superconducting contact. Nevertheless, the fact that our  $\langle G_G \rangle / \langle G_O \rangle$  is limited by the noise floor our measurement setup suggests that our semiconductor–nanowire interface homogeneity could be comparable to InAs nanowire devices using epitaxial growth techniques<sup>20</sup> or specialized surface treatments.<sup>23</sup>

We will now look at the magnetic field dependence of the hardness of the gap. We fix  $V_{BG}$  at 4.45 V and plot  $\partial I_D / \partial V_{SD}$  versus  $V_{SD}$  for several  $B_\star$  in Figure 4d. For increasing  $B_\star$ , the sharp quasiparticle peak at  $V_{SD} = 2\Delta_{Al}$  reduces in height and broadens up to  $B_{C\star,Al} \approx 300$  mT. Above  $B_{C\star,Al}$ , we enter configuration II where only X1 is superconducting but which fails to produce a clear second quasiparticle peak at  $\sim 2\Delta_{X1}$ . Instead, we see a “soft gap” signature<sup>15</sup> persisting up to  $B_{C\star,X1}$  which we attribute to X1 having an ill-defined gap due to possible diffusion-induced spatial variations in its stoichiometry or geometry.

In Figure 4e, we plot the ratio  $\langle G_G \rangle / \langle G_O \rangle$  for the three main field directions. The initial ratio is  $\sim 1 \times 10^{-3}$  in configuration I as defined in Figure 2 and the gap remains hard until we approach the critical field of Al for the respective field direction as summarized in Table 1 (see Figure S8 for the corresponding differential conductance maps for all three main field axes). The highest field where the gap remains hard,  $B_{||} \approx 250$  mT, is slightly lower than  $B_{C||,Al}$  because of the strongly reduced  $\Delta_{Al}$  at this field. The much softer gap in configuration II induced by X1 leads to a  $\langle G_G \rangle / \langle G_O \rangle \approx 1 \times 10^{-1}$  which gradually increases to 1 approaching  $B_{C,X1}$ .

Another example of the change in transport properties when Al becomes normal is seen in Figure 2a,c. Here, the fringes in the normal state attributed to MAR are only visible for  $B_\perp < B_{C\perp,Al}$ . For  $B_\perp > B_{C\perp,Al}$ , the absence of MAR suggests an increase of inelastic processes due to an ill-defined induced gap or a greatly increased quasiparticle poisoning rate.

The results in Figure 4e show that the Al contacts need to be superconducting in order to observe a hard gap. On the other hand, when only Al is superconducting, that is, going from configuration I to III, we observed no change in  $G_G$  that can be attributed to X1 becoming normal (see Figure S9 for the temperature dependence of the differential conductance at  $V_{BG} = 4.45$  V and  $B = 0$ ). This suggests that X1 does not need

to be a superconductor to observe a hard gap as long as the Al contacts proximize the entire junction. This is likely to happen, because the transparency between Al and X1 is high, and  $\Delta_{Al} > \Delta_{X1}$  indicating a coherence length for X1 comparable or larger than for Al, that is, in the order of micrometers.<sup>58</sup>

Previously, in this system a soft gap signature using NbTiN contacts has been shown<sup>33</sup> as well as a hard gap using Al contacts.<sup>35</sup> This work adds an investigation of the superconductor–semiconductor interfaces and their microscopic properties. We therefore revisit Figure 1b,c and take a closer look at the interface between the X1 and the Ge–Si island. Even though our TEM and EDX resolution prohibits a conclusive statement about the interface properties on an atomic scale, the abrupt change in contrast suggests an upper limit for the interface width of a few nanometer. As explained, this observation is supported by refs 40 and 41 showing an atomically sharp interface between the Ge and Al segment where both remain crystalline.<sup>40,41</sup> This type of interface would fit our observation of a hard gap, requiring a defect-free highly homogeneous heterointerface<sup>15</sup> and low junction transparency close to depletion. This indicates that the interdiffusion reaction between Ge and Al is essential for the observed hard superconducting gap.<sup>40,41</sup>

Utilizing these interfaces in devices suitable for measuring Majorana fermions in this system<sup>59</sup> would require a high level of control over the interdiffusion process, that is, lateral diffusion and metalization of nanowire segments should be prevented. One route would be to perform device annealing while in situ monitoring of the diffusion process as in ref 41, or possibly a higher level of control could be achieved by optimizing the annealing process. In addition, one would require thinner Al leads in order to withstand the required in-plane magnetic fields ( $>1$  T) to reach the topological phase transition.<sup>30,38</sup>

With a controlled interdiffusion reaction, the superconductors X1 and X2 themselves would also pose as interesting materials, because their high  $B_C$  in relation to their superconducting gaps might allow the creation of Majorana fermions in materials where low  $g$ -factors could be limiting.<sup>60</sup> However, more research is required to understand the soft gap induced by X1 and to fully explore the possible superconductors, their composition, and formation process.

In conclusion, we have shown that Ge–Si nanowire devices with Al contacts contain additional superconductors after annealing, caused by diffusion of Al into the nanowire channel. We identify two superconductors in two different devices: X1 is present in a Josephson FET and X2 resides in a metallic nanowire channel. Both X1 and X2 remain superconducting for magnetic fields much higher than the Al contacts which could be of potential interest for applications where proximity-induced superconductivity is required in high magnetic fields.

Close to depletion, the Josephson FET exhibits a hard superconducting gap where the in-gap conductance is suppressed by a factor  $\sim 1000$  in an SNS configuration where the in-gap conductance is close to the approximate theoretical minimum. The gap remains hard up to magnetic fields of  $\sim 250$  mT. For higher fields, a soft gap remains up to the critical field of X1. We can selectively switch Al or X1 from the normal to the superconducting state and, combined with the results of the TEM and EDX analysis, this leads us to believe that the diffusion-induced homogeneous heterointerface between the Ge core and the metalized nanowire segment is key in obtaining this hard gap. The next challenge is to more precisely



control the diffusion of Al which would grant a highly promising system for observing Majorana zero modes.<sup>30</sup>

## ■ ASSOCIATED CONTENT

### Supporting Information

The Supporting Information is available free of charge at <https://pubs.acs.org/doi/10.1021/acs.nanolett.9b03438>.

Fabrication details, specific methods used for data analysis, and additional figures (PDF)

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### Notes

The authors declare no competing financial interest.

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