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ABSTRACT

Facilitated by recent advances in strained Ge/SiGe quantum well growth technology, superconductor–semiconductor hybrid devices based on group IV materials have been developed, potentially augmenting the functionality of quantum circuits. The formation of highly transparent superconducting platinum germanosilicide (PtSiGe) contacts to Ge/SiGe heterostructures by solid-phase epitaxy between Pt and SiGe has recently been reported, although with a relatively low critical temperature <1 K. Here, we present a comparative study of the superconducting properties of Pt, Pd, Rh, and Ir germanides, along with an in-depth characterization of Ir(Si)Ge films formed by solid-phase epitaxy. For films fabricated under optimal epitaxy conditions, we report $T_c = 3.4$ K (2.6 K) for IrGe (IrSiGe). High-resolution scanning transmission electron microscopy and energy-dispersive x-ray spectroscopy reveal that Ir reacts with Ge substrates to form a polycrystalline IrGe layer with a sharp IrGe/Ge interface.

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INTRODUCTION

The development of superconductor–semiconductor hybrid technology based on group IV semiconductors^{1–11} is facilitated by advances in the growth of compressively strained Ge on Si (cs-GoS), via relaxed SiGe buffer, heterostructures, where the mobility of two-dimensional hole gases in Ge quantum wells (QWs) can exceed 10^6 cm²/(V · s).^{12–16} Aluminum forms low-resistance contacts with strained Ge due to Fermi-level pinning close to the top of the valence band,^{1,2} and superconductivity can be induced in Ge QWs by the proximity effect, either in direct Al/Ge contacts,^{3–5,10} or when Al is separated from Ge by a thin, semitransparent SiGe barrier.^{7,8} The diffusion of Al into (Si)Ge is not self-limiting, which makes the transparency of annealed Al contacts a strong function of post-deposition processing. The sensitivity of Al contacts to

thermal treatment, their low superconducting critical temperature $T_c < 1.5$ K, and low out-of-plane critical field $B_{c,\perp} \sim 10$ mT have stimulated the development of alternative contacts.

To induce a hard superconducting gap in Ge, an ideal superconducting contact should have a low Schottky barrier and form a sharp, transparent interface with Ge. In CMOS technology, mono- and polycrystalline silicides are used to form low-resistance contacts between metal interconnects and Si/Ge channels,¹⁷ in which case contacts are formed via a solid-phase reaction between a metal and Si/Ge. Although a number of silicides and germanides are superconducting at low temperatures,^{18–28} many compounds with high $T_c > 4$ K, such as NbGe₂,^{21,22,25} form at very high temperatures beyond the ~ 500 °C thermal budget of strained Ge/SiGe heterostructures.²⁹ Some platinum group metals form silicides and germanides at low temperatures (<500 °C), and the formation of superconducting PtGe

($T_c = 0.4$ K)^{18–20,23} and RhGe ($T_c = 0.96$ – 1.7 K)^{18–20,23,24} has been demonstrated. Notably, polycrystalline platinum germanosilicide (PtSiGe) contacts have been shown to induce a hard superconducting gap in shallow Ge QWs with high transparency $\tau \sim 0.95$.^{6,9} A high out-of-plane upper critical magnetic field $B_{c2,\perp} = 0.9$ T has been measured in hybrid devices with small PtSiGe leads,¹¹ substantially higher than $B_{c2,\perp} \sim 0.1$ T in large contacts.^{6,11} Another transition metal, Ta, has recently been found to form a solid solution of Ta₅Ge₃ and TaGe₂ when grown via molecular beam epitaxy (MBE) on Ge substrates at $T = 400$ °C.²⁸ These films exhibit $T_c \sim 1.8$ – 2 K, $B_{c2,\perp} \sim 1.88$ T, and $B_{c2,\parallel} \sim 5.1$ T, in agreement with previous studies of tantalum germanide.^{21,30}

Within the platinum group germanides, Ir-based compounds show the highest T_c : IrGe ($T_c = 4.7$ – 5.3 K, $B_{c1}(0) = 13.3$ mT, $B_{c2}(0) = 0.82$ – 1.13 T),^{18–20,23,24,26} Ir₃Ge₇ ($T_c = 0.87$ K),^{19,20} and IrGe₄ ($T_c = 1.22$ K, $B_{c2} = 11.5$ – 22.5 mT).²⁷ Based on results from specific heat and muon spin relaxation experiments, an enhanced T_c of IrGe is attributed to the presence of low-lying phonons.^{24,26} Although single IrGe crystals are grown at $T = 700$ °C,²⁴ there is some evidence that the onset of solid-phase reaction between Ir and Ge begins at 400 °C.³¹ While IrGe shows an enhanced T_c , IrSi is not known to be a superconductor down to mK temperatures. The superconducting properties of IrSiGe have not been investigated.

In this paper, we compare the superconducting properties of thin films formed by solid-phase epitaxy of Pd, Pt, Rh, and Ir on Ge (001) or relaxed Si_{0.15}Ge_{0.85} layers grown on Si (001) and present a systematic study of Ir-based films. We show that within the temperature range of 400 – 500 °C (within the thermal budget of strained Ge/SiGe heterostructures), self-limiting polycrystalline films of IrGe and IrSiGe are formed by solid-phase epitaxy. For optimally grown IrGe (IrSiGe) films, we report $T_c \sim 3.4$ K (2.6 K) and $B_{c2,\perp} \sim 3.4$ T (2.8 T), the latter being significantly larger than $B_c = 0.82$ – 1.13 T reported for bulk IrGe.^{24,26} High-resolution scanning transmission electron microscopy (HRSTEM) of optimally annealed Ir/Ge films reveals the formation of IrGe microcrystals with sharp boundaries between IrGe and Ge.

RESULTS

In this study, we use two types of substrates: undoped Ge (001) single-crystal wafers (*i*-Ge) and strain-relaxed Si_{0.15}Ge_{0.85} thick films capped with 2 nm of Ge, grown epitaxially on strain-relief buffers on Si (001) wafers (SiGe). All heterostructures are grown by reduced pressure chemical vapor deposition (RP-CVD).^{14,15} Prior to metal deposition, the wafers are cleaned using acetone, isopropanol, and deionized water, followed by dipping in diluted HCl and HF to remove native oxides. We expect that most of the 2 nm Ge capping layer on SiGe is naturally oxidized and is removed during the last step of acid treatment. Metal films are deposited either by DC sputtering (Ir) or electron-beam evaporation (Pt, Pd, and Rh) onto substrates held at room temperature in an ultrahigh vacuum deposition systems with a base pressure of $\sim 10^{-9}$ Torr. After metal deposition, the samples are annealed in a custom-built rapid thermal annealer (RTA) with precise temperature control, in an Ar atmosphere. Transport measurements are carried out in either a dilution refrigerator or a ³He cryostat using standard four-probe low-frequency lock-in techniques.

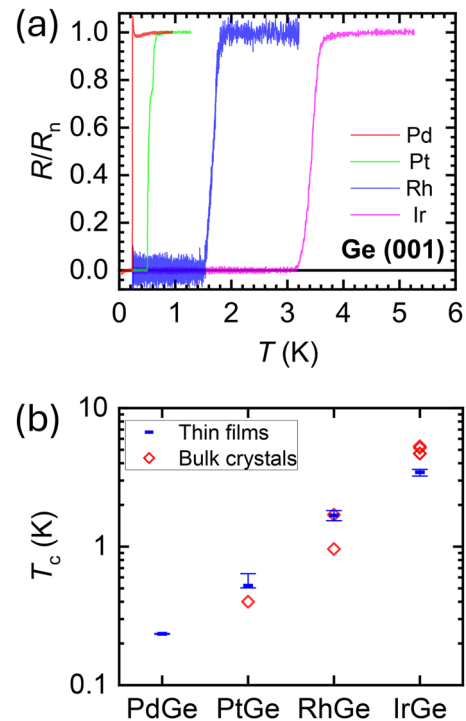


FIG. 1. (a) Temperature dependence of the normalized resistance R/R_n for optimally annealed 10 nm Pd, Pt, Rh, and Ir films deposited on Ge (001) substrates. (b) T_c for thin films in this study (blue) are compared with T_c for bulk crystals (red) from Refs. 18–20, 23, 24, and 26. The vertical bars indicate the width of the transition region defined as $0.05 < R/R_n < 0.95$.

In Fig. 1(a), the temperature dependence of the normalized resistance is plotted for Pd, Pt, Rh, and Ir germanides formed under the optimal thermal treatment conditions of 10 nm films. The superconducting transition temperatures T_c , defined here as the midpoint of the transition where the resistance drops by a factor of two relative to the normal resistance ($R = 0.5R_n$), follow the trend of bulk germanides.

Previous studies of diffusion kinetics using x-ray diffraction (XRD) and TEM techniques indicate that the solid-phase reaction between Pd and Ge initially proceeds via the diffusion-controlled growth of Pd₂Ge, which is converted into PdGe at $T > 150$ °C; PdGe phase remains stable up to its melting point $T \approx 740$ °C.^{31–42} Thus, we expect that the PdGe phase dominates in our samples for annealing at $T \geq 300$ °C, and the measured $T_c = 0.24$ K reflects the superconducting properties of PdGe films. For annealed Pt/Ge films, the measured $T_c = 0.5$ K is higher than the reported $T_c = 0.4$ K for bulk PtGe.¹⁸ Earlier studies using XRD indicate the formation of the PtGe phase when films are annealed in the 380 – 440 °C temperature range.²⁹ For Rh/Ge, we measure $T_c = 1.68$ K, close to the bulk RhGe $T_c \sim 1.7$ K.²⁴ Rh₅Ge₃ orthorhombic phase has higher $T_c = 2.12$ K,^{18–20,23} but this phase has not been found in previous studies of the solid-phase reaction of Rh/Ge thin films.^{29,31,52} RhGe₄ phase also has higher $T_c = 2.55$ K, but this phase is only stabilized under high pressure²⁷ and we do not expect it to be formed in our experiments. For Ir/Ge, the measured $T_c = 3.44$ K is slightly less than

TABLE I. Properties of optimally annealed MGe (in Fig. 1) and MSiGe films, where M = Pd, Pt, Rh, or Ir, d^M is the metal thickness, T_{ann} is the annealing temperature, and t_{ann} is the annealing time. Superconducting transition widths for critical temperature $\Delta T_c = T_c^{95} - T_c^5$ and for out-of-plane critical field $\Delta B_{c2,\perp} = B_{c2,\perp}^{95} - B_{c2,\perp}^5$, where T_c^{XX} and $B_{c2,\perp}^{XX}$ refer to temperatures and out-of-plane fields where resistance reaches XX% of its normal value. $B_{c2,\perp}(0)$ are the critical fields extrapolated to $T = 0$.

Substrate	Ge (001)				Si _{0.15} Ge _{0.85}			
M	Pd	Pt	Rh	Ir	Pd	Pt	Rh	Ir
d^M (nm)	10	10	10	10	20	20	10	10
T_{ann} (°C)	400	400	500	450	400	350	475	475
t_{ann} (min)	10	5	5	30	10	5	5	20
T_c (K)	0.235	0.522	1.68	3.44	0.299	0.585	1.28	2.56
ΔT_c (K)	0.002	0.134	0.28	0.38	0.01	0.02	0.22	0.31
$B_{c2,\perp}(0)$ (T)	0.007	0.11	1.46	3.43	...	0.18	1.37	2.84
$\Delta B_{c2,\perp}$ (T) ^a	0.001	0.06	0.30	1.38	...	0.12	0.15	0.73

^aMeasured at $T = 0.25$ K for RhGe, IrGe, and IrSiGe films and $T \sim 50$ mK for all others films.

the 4.7–5.3 K reported for stoichiometric IrGe crystals^{18,24} but higher than that for Ir₃Ge₇ ($T_c = 0.87$ K¹⁹) or IrGe₄ ($T_c = 1.22$ K²⁷) phases.

The superconducting properties of Pd, Pt, Rh, and Ir germanosilicides in Table I are found to reflect the composition of the Si_xGe_{1-x} substrate. For films grown on Si_{0.15}Ge_{0.85} substrates, we measure $T_c = 0.30$ K for Pd/SiGe, $T_c = 0.59$ K for Pt/SiGe, $T_c = 1.28$ K for Rh/SiGe, and $T_c = 2.56$ K for Ir/SiGe. These values are close to $T_c^{\text{MSiGe}} \approx 0.15 \times T_c^{\text{Si}} + 0.85 \times T_c^{\text{MGe}}$ (M = Pd, Pt, Rh, or Ir) (Fig. S1 in the [supplementary material](#)). The observation of a weighted T_c suggests the formation of a proportional mixture of germanides and silicides.

The dependence of T_c and B_{c2} (defined at a midpoint of resistance drop, $R(B_{c2}) = 0.5R_n$) of Ir/Ge and Ir/SiGe films on Ir thickness, annealing time t_{ann} , and annealing temperature is summarized in Fig. 2. The initial increase in T_c and $B_{c2,\perp}$ with t_{ann} is attributed to the finite barrier for Ir diffusion into *i*-Ge and SiGe

substrates. The corresponding activation energy can be found by analyzing the time t_{ann}^0 needed to anneal a film to reach the same T_c for different annealing temperatures T_{ann} . The $t_{\text{ann}}^0(T_{\text{ann}})$ dependence for $T_c \sim (2.8 \pm 0.2)$ K is found to be exponential, and from the Arrhenius plot, we extract the activation energy $E_a \approx 2.7$ eV assuming $t_{\text{ann}}^0 \propto 1/D$ and a diffusion coefficient $D \propto \exp(-E_a/k_B T_{\text{ann}})$, where k_B is the Boltzmann constant (Fig. S2 in the [supplementary material](#)). Here, we assume that partially formed germanides are morphologically similar and neglect the reduction in T_c due to the reverse proximity effect in the presence of metallic Ir in under-annealed samples. The samples with thinner Ir films require shorter annealing time to reach the same T_c .

For Ir on *i*-Ge samples, after an initial increase from $T_c \sim 0.2$ K for Ir films with annealing time, T_c reaches a maximum and then decreases with further annealing. A sharp decrease in T_c is observed for samples annealed at $T \geq 475$ °C, where an extra minute

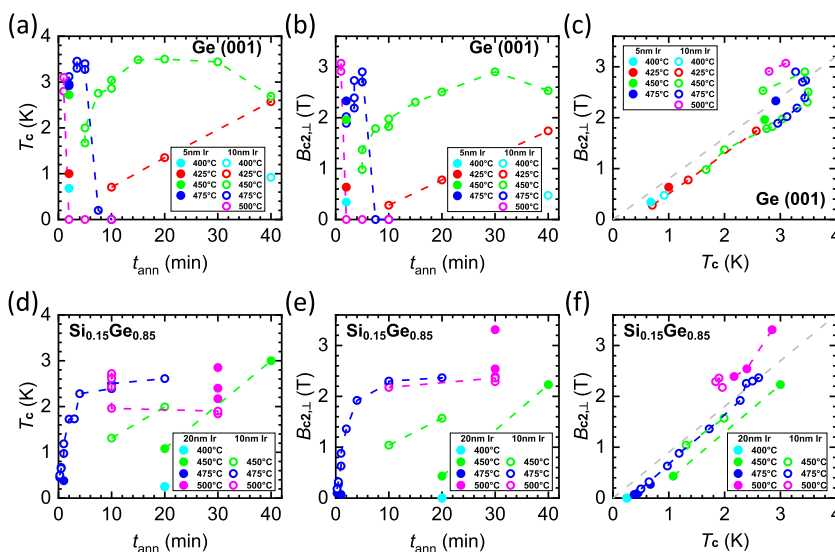


FIG. 2. [(a), (b), (d), and (e)] The critical temperature T_c and the out-of-plane critical field $B_{c2,\perp}$ are plotted as a function of annealing time t_{ann} for *i*-Ge and SiGe substrates, showing their dependence on the Ir film thickness and annealing temperature. $T_c < 0.25$ K is plotted as $T_c = 0$. The filled (open) circles correspond to different Ir thicknesses indicated in the legends, and the colors differentiate different annealing temperatures. [(c) and (f)] $B_{c2,\perp}$ is re-plotted as a function of T_c .

of annealing results in a drop in T_c from 3.2 K to <0.25 K. For IrGe films, the optimal annealing temperature is found to be 450 °C; this T_{ann} provides a large $t_{\text{ann}} \approx 12\text{--}30$ min window to form films with the highest T_c . As we will discuss below, optimally annealed films consist of IrGe microcrystals.

For Ir on SiGe, no significant decrease in T_c for long annealing times has been observed. We speculate that the reduction in T_c in over-annealed Ir/Ge samples is due to the formation of a competing semiconducting Ir_4Ge_5 phase with lower formation energy.^{43–45} This explanation is consistent with the observed higher stability of IrSiGe films, since IrSi is expected to have a lower formation energy than the competing Ir_3Si_4 and Ir_3Si_5 metallic phases.^{43–45} The formation of a semi-insulating phase in over-annealed Ir/Ge samples is also consistent with a sharp increase in resistivity and a semiconducting T -dependence (slight resistance increase below 1 K) (Table S1 and Fig. S3 in the [supplementary material](#)).

The critical field $B_{c2,\perp}$, plotted in Fig. 2(b) and 2(e), evolves similarly to T_c as a function of Ir thickness, annealing time, and annealing temperature. This similarity is emphasized by an almost linear dependence $B_{c2,\perp} \propto T_c$, shown in Fig. 2(c) and 2(f), where $B_{c2,\perp}$ is plotted as a function of T_c for all devices and annealing conditions. The highest $B_{c2,\perp} = 3.07$ T measured in Ir/Ge and $B_{c2,\perp} = 3.31$ T measured in Ir/SiGe films are significantly higher than the $B_{c2,\perp} = 0.82\text{--}1.13$ T reported for bulk IrGe crystals.^{24,26}

In Fig. 3, the temperature dependence of resistivity is plotted for different in-plane ($B_{\parallel}$) and out-of-plane ($B_{\perp}$) magnetic fields. In the insets, the temperature dependencies of $B_{c2,\perp}$ and $B_{c2,\parallel}$ are fitted to

$$B_{c2}(T) = B_{c2}(0) \left(1 - \frac{T}{T_c}\right)^{\alpha}, \quad (1)$$

where α and $B_{c2}(0)$ are the fitting parameters summarized in Table II. For the out-of-plane field $\alpha \approx 1$, while for the in-plane

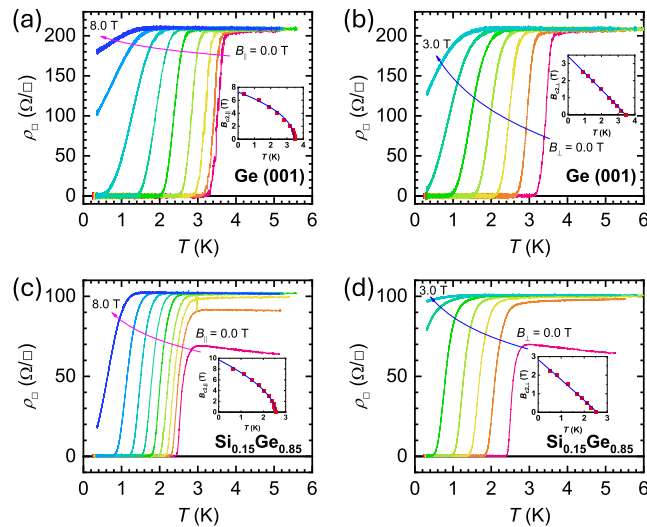


FIG. 3. The change in resistivity as a function of temperature is plotted for various [(a) and (c)] in-plane $B_{\parallel}$ and [(b) and (d)] out-of-plane $B_{\perp}$ magnetic fields for the optimally annealed [(a) and (b)] Ir/Ge and [(c) and (d)] Ir/SiGe films. The insets show the temperature dependence of B_{c2} , and the blue lines represent fits using Eq. (1).

TABLE II. T_c is an experimental value for the films studied in Fig. 3, while B_{c2} and α are extracted from the fits to Eq. (1). B_P is an estimated Pauli limit as discussed in the text. The superconducting coherence length $\xi(0)$ and effective film thickness d_{eff} are calculated from measured in-plane (IP) and out-of-plane (OOP) critical fields $B_{c2,\perp}(T) = \Phi_0/(2\pi\xi^2)$ and $B_{c2,\parallel}(T) \approx \sqrt{3}\Phi_0/(\pi\xi d_{\text{eff}})$. The Ginzburg–Landau coherence length ξ_0 and mean free path l are calculated using $\xi(0) = \sqrt{\xi_0 l}$ and $l = \hbar^3 \sqrt{3\pi^2} n^{2/3} / (e^2 \rho)$, where n is the carrier density, ρ is the three-dimensional resistivity, and $\xi(0)$ is ξ at the lowest T .

	Ir/Ge		Ir/SiGe	
	IP	OOP	IP	OOP
T_c (K)	3.44		2.56	
$B_{c2}(0)$ (T)	7.28	3.43	9.71	2.84
α	0.46	0.97	0.57	0.98
B_P (T)	8.3		6.2	
$\xi(0)$ (nm)	9.8		10.8	
d_{eff} (nm)	16.0		10.9	
l (nm)	1.7		...	
ξ_0 (nm)	56.5		...	

field $\alpha \approx 0.5$, confirming the two-dimensional nature of IrGe and IrSiGe films with $d_{\text{eff}} \lesssim 1.8\xi$, where d_{eff} is the effective film thickness and ξ is the superconducting coherence length, extracted from the temperature dependence of the critical fields, $B_{c2,\perp}(T) = \Phi_0/(2\pi\xi^2)$ and $B_{c2,\parallel}(T) \approx \sqrt{3}\Phi_0/(\pi\xi d_{\text{eff}})$. This temperature dependence originates from the temperature dependence of the coherence length, $\xi(T) \approx \xi(0)/\sqrt{1 - T/T_c}$, where in a dirty superconductor $\xi(0) = \sqrt{\xi_0 l}$ is a geometrical average of a Ginzburg–Landau coherence length ξ_0 and a mean free path l , $\Phi_0 = h/2e$ is the magnetic flux quantum.⁴⁶ The mean free path, $l = \hbar^3 \sqrt{3\pi^2} n^{2/3} / (e^2 \rho)$, is calculated using the resistivity ρ and electron density n measured in the normal state just above T_c . The ratio $\xi_0/l = 33.2$ indicates dirty-limit superconductivity in IrGe. We note that the critical in-plane field is comparable to (IrGe), or exceeds (IrSiGe), the Pauli limit $B_P = \Delta_0/(\sqrt{2}\mu_B)$, where $\Delta_0 \approx 2.3k_B T_c$ is the zero-temperature superconducting gap for Ir(Si)Ge and μ_B is the Bohr magneton, indicating strong spin–orbit interactions.⁴⁷ The high $B_{c2,\perp}$ values, $B_{c2,\perp}(\text{IrGe}) = 3.43$ T and $B_{c2,\perp}(\text{IrSiGe}) = 2.84$ T, significantly exceed the 1.13 T reported in bulk IrGe.²⁶ This enhancement can be attributed to the microcrystalline nature of the films.⁴⁸ These high $B_{c2,\perp}$ values could be useful for studying the interplay between superconductivity and quantum Hall states in high-mobility Ge two-dimensional hole gases.

The high-resolution scanning transmission electron microscopy analysis of a 10 nm Ir/i-Ge film annealed at 475 °C for 3.5 min ($T_c \sim 3.4$ K) is presented in Figs. 4 and 5. Low-resolution image Fig. 4(a) shows the formation of a uniform film with a thickness of $d = (24 \pm 3)$ nm. The density of IrGe, 13.10 g/cm³, can be calculated from its structural parameters.^{24,26} Assuming that all Ir reacts with Ge to form the IrGe phase, 10 nm of Ir should react with 16 nm of Ge to form 23.7 nm of IrGe. This value is in excellent agreement with the film thickness d obtained from the wide-area STEM image

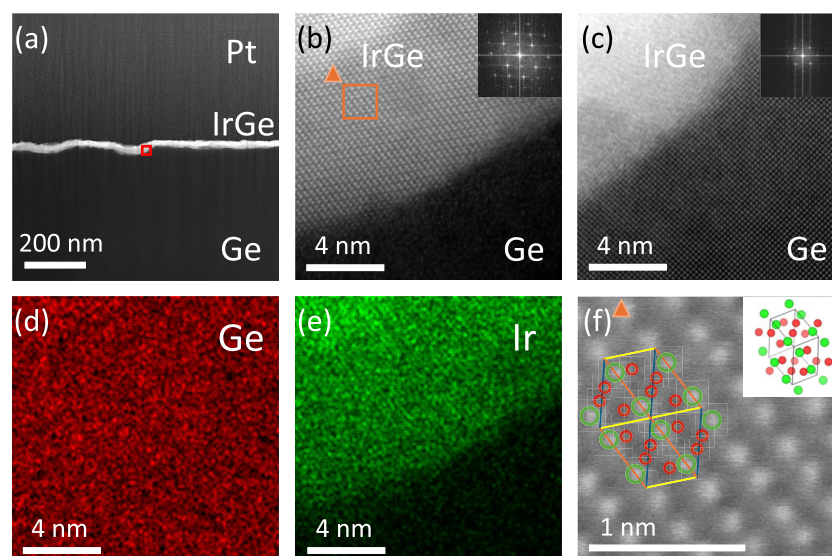


FIG. 4. (a) Low magnification cross-sectional STEM image of an Ir 10 nm/Ge (001) sample annealed at 475 °C for 3.5 min. The sample is coated with Pt protection layer. [(b) and (c)] High-angle annular dark-field scanning transmission electron microscopy (HAADF-STEM) near IrGe/Ge interface [red region in (a)] is performed at slightly different angles to match either [111] orientation of IrGe or [100] orientation of Ge lattices. Insets: fast Fourier transforms (FFT) of the images. [(d) and (e)] Energy-dispersive x-ray spectroscopy (EDX) of (b). (f) Atomic resolution STEM image of IrGe [orange region in (b)] with red and green circles indicating the positions of Ir and Ge atoms correspondingly. The yellow, blue, and orange lines indicate the *a*, *b*, and *c* axes of the IrGe lattice. Inset: a view of an IrGe lattice seen along [111] direction.

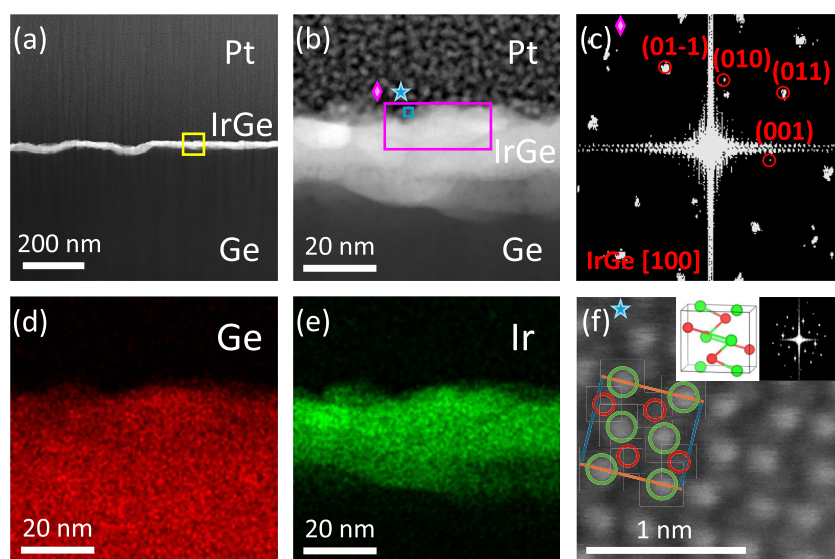


FIG. 5. (a) Low-magnification cross-sectional STEM image of an Ir 10 nm/i-Ge film annealed at 475 °C for 3.5 min. The sample is coated with a Pt protection layer. (b) HAADF-STEM image of a flat region of the IrGe film [yellow region in (a)], with (c) the indexed FFT [of the magenta region in (b)] and [(d) and (e)] corresponding EDX maps. (f) Atomic-resolution STEM image near the top surface of the film [blue region in (b)], showing the orthorhombic phase (Pnma, space group No. 62) of IrGe, taken along the [100] orientation of the IrGe grain. Insets: schematic of the IrGe lattice and FFT of (f). The green circles mark Ir atoms and the red circles mark Ge atoms. The *b* and *c* axes of the IrGe lattice are highlighted by blue and orange lines, respectively.

Fig. 4(a), suggesting that most Ir reacts with Ge to form IrGe, and close to the effective thickness d_{eff} calculated in Table II.

This conclusion is corroborated by cross-sectional aberration-corrected high-angle annular dark-field scanning transmission electron microscopy (HAADF-STEM) images, which show the formation of IrGe microcrystals with lateral sizes of >10 nm throughout the thickness of the film. In Fig. 4(b) and 4(c), an IrGe/Ge boundary is shown at two slightly different angles, highlighting the crystalline structure of an IrGe crystal and the surrounding Ge matrix. The

boundary is sharp, with a negligibly small concentration of Ir in Ge, as evidenced by the energy-dispersive x-ray spectroscopy (EDX) analysis of Ir and Ge distribution across the boundary Figs. 4(d), 4(e), 5(d), and 5(e). Note that the STEM specimen is $\lesssim 100$ nm thick, a few times larger than the average IrGe grain size. Unlike STEM, which probes only a few nanometers from the surface, EDX averages the Ir concentration over the entire thickness of the film. Near an IrGe/Ge interface EDX may pick up a signal from an overlapping IrGe microcrystal at the back of the specimen not visible in the

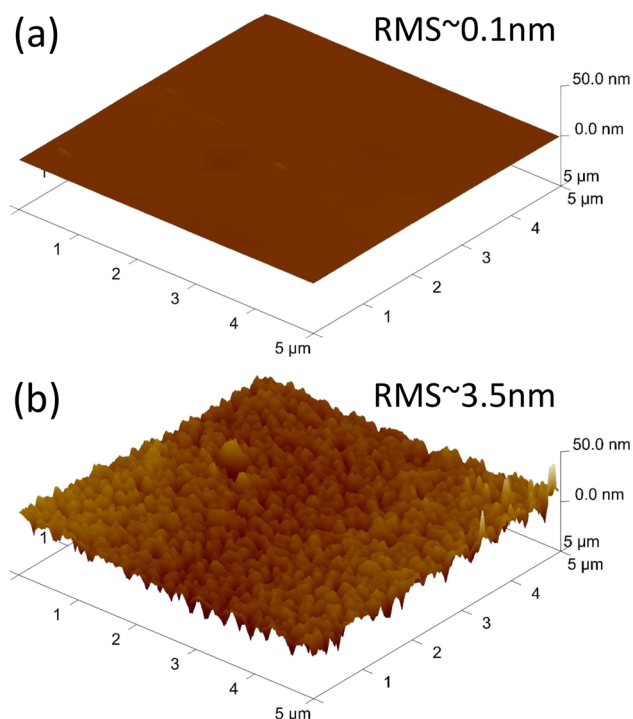


FIG. 6. AFM images of 10 nm Ir on *i*-Ge before (a) and after (b) annealing at 475 °C for 3.5 min.

STEM image, which can explain some small Ir background detected in the otherwise stoichiometrically perfect Ge matrix.

The detailed analysis of an atomic-resolution image of IrGe microcrystals Fig. 5(f) reveals the formation of an orthorhombic IrGe phase (Pnma, space group No. 62) with lattice parameters $b = (5.607 \pm 0.028)$ Å, $c = (6.294 \pm 0.018)$ Å, and angle $(89.62 \pm 0.34)^\circ$ between b and c . Lattice parameters determined for bulk IrGe crystals using XRD^{24,26} fall within the error bars of our measurements, indicating the formation of unstrained IrGe.

The surface morphology studies of IrGe films corroborate the formation of a polycrystalline film. In Fig. 6, we plot atomic force microscope (AFM) images of a sample before and after annealing. The surface of an Ir/Ge (001) sample before annealing is smooth with an RMS roughness $R_q \sim 0.1$ nm. Some shallow holes (a few nm deep) likely originate from the surface of the Ge wafer and formed during acid cleaning prior to Ir deposition. The surface of the annealed sample shows the formation of bumps with a lateral size (57 ± 22) nm and $R_q \sim (3.5 \pm 1.0)$ nm. Based on TEM images, these bumps can be attributed to the formation of IrGe microcrystals.

DISCUSSION

In this paper, we have investigated and compared the superconducting properties of Pd, Pt, Rh, and Ir germanide thin films formed by solid-phase epitaxy, with the detailed characterization of IrGe and IrSiGe films. The optimal annealing temperature increases from Pd to Ir but remains within the thermal budget of strained Ge/SiGe heterostructures. We verified that the quantum mobility

$\mu_q = (3 \sim 5) \times 10^4 \text{ cm}^2/(\text{V} \cdot \text{s})$ of the two-dimensional hole gas in strained Ge quantum wells is not substantially degraded by the highest annealing temperatures used in this study. The high-resolution STEM and EDX analyses of IrGe films reveal that Ir reacts with Ge to form a self-limiting, polycrystalline IrGe layer with a sharp IrGe/Ge interface. Optimally annealed IrGe films exhibit the highest critical temperature $T_c \approx 3.4$ K and upper critical magnetic field $B_{c2} \approx 3.4$ T. In contrast, over-annealed IrGe films show a suppression of superconductivity, with both T_c and B_{c2} dropping to near zero, which we attribute to the formation of the semiconducting Ir_4Ge_5 phase. IrSiGe films exhibit slightly lower superconducting parameters, with $T_c \approx 2.6$ K and $B_{c2} \approx 2.8$ T but demonstrate greater thermal stability. This enhanced robustness is likely due to the formation of the more stable metallic IrSi phase. Our results establish that Ir(Si)Ge thin films formed via solid-phase epitaxy provide a viable route for fabricating high- T_c , high- B_c superconducting contacts to Ge-based Josephson field-effect transistors (Ge JoFETs). These materials offer a promising platform for integrating high-performance superconducting electronics with strained Ge quantum wells in a process fully compatible with mature CMOS technology.

SUPPLEMENTARY MATERIAL

The [supplementary material](#) contains detailed information on sample fabrication and characterization methods; tables that summarize the dependence of films properties on annealing conditions; the additional figures of transport analysis of annealed germanide or germanosilicide films; the dependence of transport properties of strained Ge quantum wells on annealing conditions; and the methods for the analysis of IrGe lattice constants and microcrystal sizes.

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AUTHOR DECLARATIONS

Conflict of Interest

The authors have no conflicts to disclose.

Author Contributions

Hao Li: Conceptualization (equal); Data curation (lead); Formal analysis (lead); Investigation (lead); Validation (lead);

Visualization (lead); Writing – original draft (lead); Writing – review & editing (equal). **Zhongxia Shang**: Investigation (equal); Writing – review & editing (supporting). **Michael P. Lilly**: Resources (equal); Writing – review & editing (supporting). **Maksym Myronov**: Resources (equal); Writing – review & editing (supporting). **Leonid P. Rokhinson**: Conceptualization (lead); Formal analysis (equal); Funding acquisition (lead); Methodology (lead); Project administration (lead); Supervision (lead); Visualization (equal); Writing – original draft (equal); Writing – review & editing (lead).

DATA AVAILABILITY

The data that support the findings of this study are available within the article and its [supplementary material](#).

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Characterization of superconducting germanide and germanosilicide films of Pd, Pt, Rh and Ir formed by solid-phase epitaxy

Supplementary Material

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The supplementary material contains detailed information on sample fabrication and characterization methods; tables that summarize the dependence of films properties on annealing conditions; additional figures of transport analysis of annealed germanide or germanosilicide films; the dependence of transport properties of strained Ge quantum wells on annealing conditions; and the methods for the analysis of IrGe lattice constants and microcrystal sizes.

I. METHODS

$\text{Si}_{0.15}\text{Ge}_{0.85}$ wafers used in this study were grown on *p*-doped Si (001) substrates by reduced-pressure chemical vapor deposition (RP-CVD). Undoped Ge (001) wafers were purchased from MTI Corporation. All wafers were degreased using toluene, acetone, and isopropanol with ultrasonication. Prior to loading into the deposition system, the native surface oxide was removed by sequential dips in $\text{HCl}:\text{H}_2\text{O}$ (1:6) for 15 seconds and $\text{HF}:\text{H}_2\text{O}$ (1:10) for 10 seconds, followed by a 10-second rinse in deionized (DI) water.

Iridium (Ir) was deposited by DC sputtering in a KJL PVD-75 system at Sandia National Laboratories under a 3 mTorr Ar atmosphere, using 100 W DC power, substrate rotation at 20 rpm, and a 75° tilt angle, at a deposition rate of approximately 1 Å/s. Palladium (Pd), platinum (Pt), and rhodium (Rh) films were deposited at normal incidence by electron-beam evaporation in an AJA ATC 1800-HY hybrid deposition system at Purdue University, with a base pressure below 10^{-8} Torr and a deposition rate of 1–1.5 Å/s. In both systems, substrates were maintained at room temperature during deposition.

Following deposition, samples were annealed in an Ar atmosphere using a home-built rapid thermal annealing system with a heating and cooling rate of $1^\circ\text{C}/\text{s}$. Electrical and magnetotransport measurements were conducted on samples patterned in either Hall bar or van der Pauw geometries, using a ^3He system or a dilution refrigerator. Conventional low-frequency (10–30 Hz) four-terminal lock-in techniques were employed.

For high-resolution scanning transmission electron microscopy (HRSTEM) studies, a < 100 nm-thick lamella was prepared using a Thermo Scientific Helios G4 UX DualBeam focused ion beam (FIB) system. Transmission electron microscopy (TEM) imaging and energy-dispersive X-ray spectroscopy (EDX) were performed using a Thermo Scientific Themis Z system at an acceleration voltage of 300 kV. Atomic force microscopy (AFM) images were

acquired using a Veeco Dimension 3100 AFM system.

II. EXTENDED TABLES

TABLE S1. Parameters of IrGe (top) and IrSiGe (bottom) films formed using different annealing conditions. Most parameters are defined in Table. I in the main text. Sheet resistivity $\rho_{\square}$ is measured in van der Pauw geometry. Resistivity ρ and electron density n are calculated assuming that all Ir reacts with Ge to form IrGe with the thickness $2.37 d^{\text{Ir}}$.

d^{Ir} (nm)	T_{ann} (°C)	t_{ann} (min)	T_{c} (K)	ΔT_{c} (K)	$B_{\text{c}2,\perp}$ (T)	$\Delta B_{\text{c}2,\perp}$ (T)	$\rho_{\square}$ ($\Omega/\square$)	ρ ($\mu\Omega\text{-m}$)	n 10^{27}m^{-3}
i-Ge	400	2	0.68	0.09	0.34	0.16	136	—	—
	425	2	1.00	0.08	0.64	0.17	134	—	—
	450	2	2.72	0.14	1.96	0.33	117	1.39	—
	475	2	2.92	0.16	2.33	0.46	115	1.36	—
	350	40	0.39	0.04	0.12	0.11	38.4	—	—
	375	40	0.35	0.02	0.10	0.02	40.0	—	—
	400	40	0.92	0.07	0.48	0.10	51.3	—	—
	450	30 ^{a)}	3.44	0.31	2.90	1.39	209	4.95	1.88
	475	3.5	3.30	0.23	2.19	0.58	44.5	1.05	13.9
	475	3.5	3.44	0.19	2.39	0.76	52.0	1.23	—
	500	1	3.10	0.84	3.07	2.10	217	5.14	—
	500	2 ^{b)}	<0.25	—	—	—	430	—	—
	450	20	1.99	0.23	1.57	0.45	64.9	1.54	—
	475	10	2.50	0.21	2.30	0.65	59.5	1.41	—
	475	20 ^{a)}	2.61	0.32	2.37	0.76	64.2	1.52	—
SiGe	450	20	1.99	0.23	1.57	0.45	64.9	1.54	—
10	475	10	2.50	0.21	2.30	0.65	59.5	1.41	—
	475	20 ^{a)}	2.61	0.32	2.37	0.76	64.2	1.52	—

^{a)} These conditions are used in the main text analysis.

^{b)} For this annealing condition, there is an onset of a broad transition with $T_{\text{c}}^{95} = 1.00$ K and $B_{\text{c}2,\perp}^{95} = 2.71$ T.

TABLE S2. Parameters of some PdGe films formed using different annealing conditions. Most parameters are defined in Table. I in the main text. Sheet resistivity $\rho_{\square}$ is measured in van der Pauw geometry. Resistivity ρ is calculated assuming that all Pd reacts with Ge to form PdGe with the thickness $2.15 d^{\text{Pd}}$.

d^{Pd}	T_{ann}	t_{ann}	T_{c}	ΔT_{c}	$B_{\text{c}2,\perp}$	$\Delta B_{\text{c}2,\perp}$	$\rho_{\square}$	ρ
(nm)	(°C)	(min)	(K)	(K)	(T)	(T)	($\Omega/\square$)	($\mu\Omega\text{-m}$)
10	350	10	0.236	0.043	0.005	0.026	4.29	0.09
	400	10	0.235	0.002	0.007	0.001	5.30	0.11

TABLE S3. Parameters of some RhSiGe films formed using different annealing conditions. Most parameters are defined in Table. I in the main text. Sheet resistivity $\rho_{\square}$ is measured in van der Pauw geometry. Resistivity is calculated assuming that all Rh reacts with Ge to form RhGe with the thickness $2.16 d^{\text{Rh}}$.

d^{Rh} (nm)	T_{ann} (°C)	t_{ann} (min)	T_{c} (K)	ΔT_{c} (K)	$B_{\text{c}2,\perp}$ (T)	$\Delta B_{\text{c}2,\perp}$ (T)	$\rho_{\square}$ ($\Omega/\square$)	ρ ($\mu\Omega\text{-m}$)
10	450	10	1.26	0.03	0.98	0.40	17.1	0.37
	475	5	1.28	0.22	1.30	0.15	25.1	0.54
	500	5	1.26	0.02	1.06	0.60	22.4	0.48

III. EXTENDED FIGURES

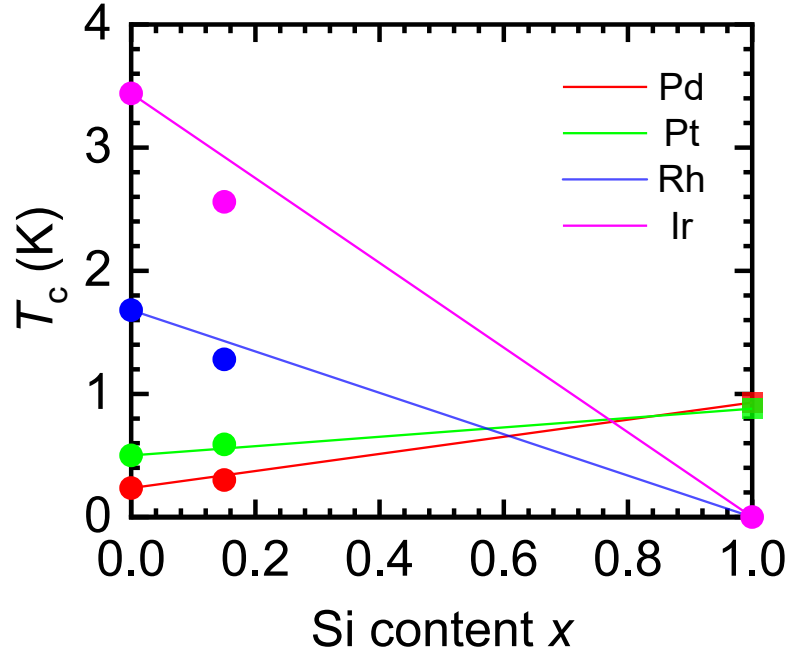


FIG. S1. T_c for pure MSi are shown by squares symbols¹⁻⁴. Measured T_c for MGe, MSi and MSi_{0.15}Ge_{0.85} are plotted as dots. These values are close to the values linearly extrapolated between T_c of MSi and MGe.

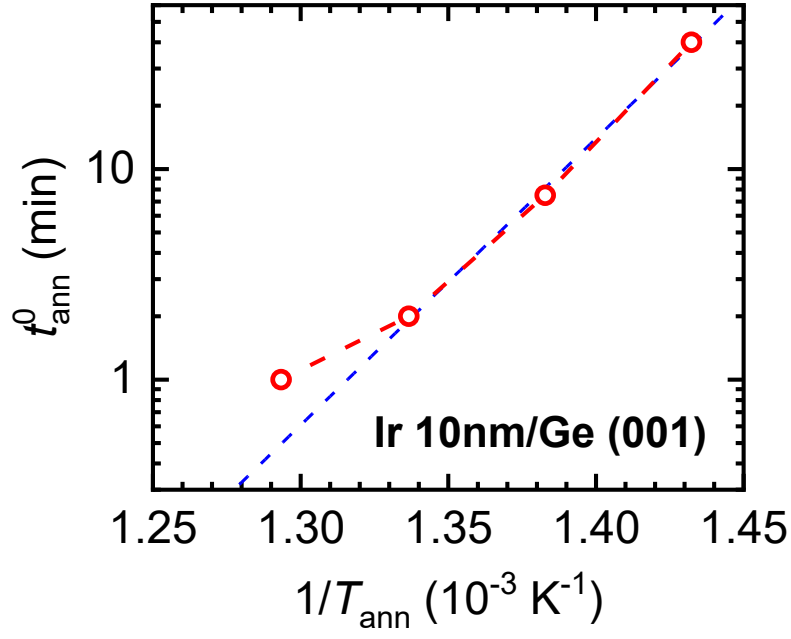


FIG. S2. The Arrhenius plot of t_{ann}^0 vs. $1/T_{\text{ann}}$ for Ir 10 nm/*i*-Ge films. t_{ann}^0 is defined as the annealing time required for the film to have a critical temperature $T_c \sim (2.8 \pm 0.2)$ K. The dashed line corresponds to the activation energy 2.7 eV.

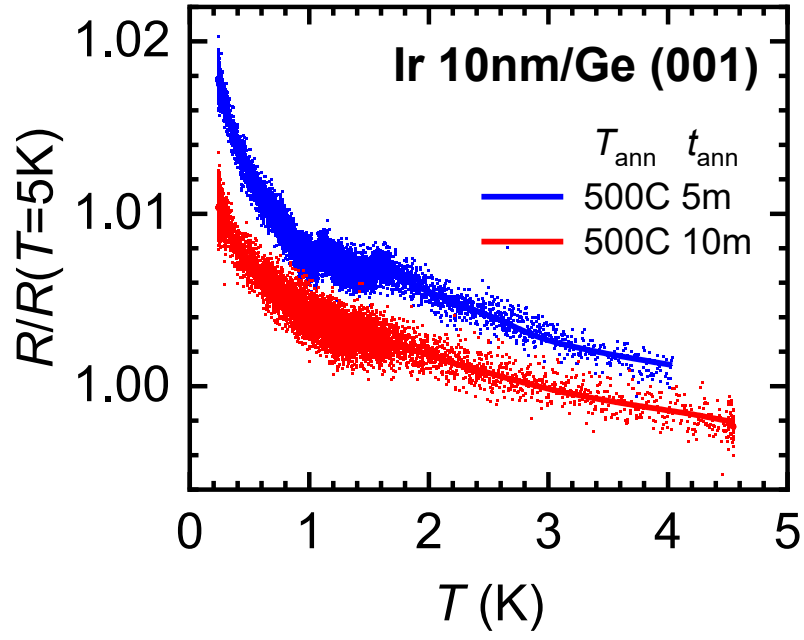


FIG. S3. Temperature dependence of normalized resistance in over-annealed Ir/*i*-Ge films shows a negative trend at low temperatures, indicating semiconducting properties of the film.

IV. THERMAL BUDGET OF STRAINED GE QUANTUM WELLS

For superconducting germanides to be used with strained Ge quantum wells grown at $\sim 550^\circ\text{C}$, it is important to identify annealing conditions beyond which the quality of the two-dimensional (2D) gas is degraded, either due to strain relief or Si diffusion across the SiGe/Ge interface. As a metric, we choose the quantum mobility of a 2D hole gas $\mu_q = 1/B^*$, which can be obtained from the onset magnetic field B^* where Schubnikov-de Haas (SdH) oscillations appear. For these experiments we used a deep (200 nm) quantum well with modulation doping, where density and mobility are not very sensitive to the surface condition and 2D gas with hole density $\sim 1.2 \cdot 10^{11} \text{ cm}^{-2}$ is formed in as-grown wafer without external electric field. The samples were annealed at T_{ann} for t_{ann} in a forming gas, then indium contacts were made to the corners of square samples in order to measure transport properties in the van der Paw geometry. Transport measurements were performed at $T = 250 \text{ mK}$ using low frequency lock-in techniques.

In Table. S4, we show that wafers annealed at 475°C do not degrade at least up to 20 min of annealing. A sample annealed at 500°C for 10 minutes shows reduced μ_q indicating degradation of the 2D gas quality either due to the reduction of the quantum well depth due to partial Ge strain relieve, or the onset of Si migration into the quantum well. Therefore, we use 500°C as the upper limit for our studies.

TABLE S4. Dependence of a 2D hole gas quantum mobility μ_q on the annealing temperature T_{ann} and time t_{ann} .

T_{ann} ($^{\circ}\text{C}$)	475						500
t_{ann} (min)	1	2	4	10	10	20	10
p ($10^{11}/\text{cm}^2$)	1.21	1.17	1.22	1.12	1.55	1.41	1.21
B^* (T)	0.20	0.22	0.21	0.20	0.21	0.20	0.30
μ_q ($10^4 \text{ cm}^2/(\text{V} \cdot \text{s})$)	5.0	4.5	4.8	5.0	4.8	5.0	3.3

V. ANALYSIS OF IRGE LATTICE CONSTANTS AND MICROCRYSTAL SIZES

The lattice constants of IrGe microcrystals were measured and calculated from STEM images. In high-magnification STEM images, such as Fig. 5(f) in the main text, the periods of Ir lattices along different orientations are directly measured using ImageJ software⁵. Using the calibrations derived from Ge lattices, such as Fig. 4(c) in the main text, which has known lattice constants, the lattice constants of IrGe crystals are calculated and averaged over several images.

The size of IrGe microcrystals was estimated from STEM and AFM images. In low-magnification STEM images, such as Fig. 4(b) in the main text, grain boundaries are visible, and grain sizes are measured directly using ImageJ software⁵ and averaged over several grains in the image. AFM images are analyzed using Gwyddion software⁶, which calculates the average grain size and the grain size distribution.

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