Superconductivity in rhombohedral trilayer graphene

Haoxin Zhou1, Tian Xie1, Takashi Taniguchi2, Kenji Watanabe3 & Andrea F. Young1✉

To access superconductivity via the electric field effect in a clean, two-dimensional device is a central goal of nanoelectronics. Recently, superconductivity has been realized in graphene moiré heterostructures1–4; however, many of these structures are not mechanically stable, and experiments show signatures of strong disorder. Here we report the observation of superconductivity—manifesting as low or vanishing resistivity at sub-kelvin temperatures—in crystalline rhombohedral trilayer graphene5,6, a structurally metastable carbon allotrope. Superconductivity occurs in two distinct gate-tuned regions (SC1 and SC2), and is deep in the clean limit defined by the ratio of mean free path and superconducting coherence length. Mapping of the normal state Fermi surfaces by quantum oscillations reveals that both superconductors emerge from an annular Fermi sea, and are proximal to an isospin-symmetry-breaking transition where the Fermi surface degeneracy changes7. SC1 emerges from a paramagnetic normal state, whereas SC2 emerges from a spin-polarized, valley-unpolarized half-metal17 and violates the Pauli limit for in-plane magnetic fields by at least one order of magnitude8,9. We discuss our results in view of several mechanisms, including conventional phonon-mediated pairing10,11, pairing due to fluctuations of the proximal isospin order12, and intrinsic instabilities of the annular Fermi liquid13,14. Our observation of superconductivity in a clean and structurally simple two-dimensional metal provides a model system to test competing theoretical models of superconductivity without the complication of modelling disorder, while enabling new classes of field-effect controlled electronic devices based on correlated electron phenomena and ballistic electron transport.

Owing to the instability of Fermi liquids to arbitrarily weak attractive interactions15, most elemental metals become superconducting at sufficiently low temperatures when the thermal fluctuation is smaller than the attractive interactions. However, some metals become magnetic instead. In these systems, time reversal symmetry is spontaneously broken, suppressing conventional superconducting pairing that relies on the degeneracy of Kramers pairs. The competition between magnetism and superconductivity can be understood from the point of view of the density of states. High density of states simultaneously favours superconductivity10 and magnetism16, with the ground state determined by the relative strength of the effective attractive interaction (typically mediated by phonons) and inter-electron Coulomb repulsion. In other situations (for example, in heavy-fermion compounds15), magnetism and superconductivity may be cooperative. In this scenario, magnetic fluctuations may themselves mediate attractive interactions between electrons10, typically resulting in superconductivity with pairing symmetries other than s-wave.

Here we report the discovery of superconductivity in rhombohedral trilayer graphene (RTG) on the cusp of an isospin-symmetry-breaking transition. The crystal structure of RTG is shown in Fig. 1a. As in other honeycomb carbon systems, near zero doping the Fermi surfaces are localized to the two inequivalent valleys at the corners of the hexagonal Brillouin zone. Of relevance for isospin symmetry breaking, the valley provides an internal degree of freedom in addition to the electron spin. In the absence of an applied perpendicular displacement field \( D \), the noninteracting electronic structure of RTG is described by three Dirac crossings in each valley (Fig. 1b). At finite carrier density \( n_e = 10^{12}\text{cm}^{-2} \), these Dirac pockets merge at a saddle-point van Hove singularity where the density of states diverges (Fig. 1c, d). At finite \( D \) the Dirac cones become gapped, and the van Hove singularities are enhanced in magnitude5,6. Experimentally, RTG hosts a cascade of transitions at finite doping18 where one or more of the spin and valley symmetries spontaneously breaks. These instabilities appear to be generic to rhombohedral graphite19, and are predicted to apply to Bernal bilayer graphene as well19.

Superconducting phenomenology in RTG

Our main result is summarized in Fig. 1e, which shows a false-colour plot of the longitudinal resistivity \( R_{xx} \) as a function of \( D \) and \( n_e \). We observe two distinct superconducting states at these densities, which render in bright cyan on the colour scale off Fig. 1e and which we denote

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SCI and SC2. Both states show nonlinear transport signatures typical of superconductivity at sufficiently low temperatures (Fig. 1f, g and additional data in Extended Data Fig. 1). Fitting the nonlinear voltage to a Berezinskii–Kosterlitz–Thouless model22 gives \( T_{\text{FBT}} = 106 \text{ mK} \) for SCI, while for SC2 \( T_{\text{FBT}} \) appears to be just below the base temperature of our measurement system (Extended Data Fig. 2).

Notably, both superconducting states occur near transitions in the normal state resistivity associated with a change in the degeneracy of the Fermi surface8; in other words, superconductivity occurs at a symmetry breaking transition. To better understand this connection, we measure quantum oscillations at low magnetic fields \( B < 1 \) T (Fig. 2a) in the density range spanning SCI at fixed \( D = 0.4 \text{ V nm}^{-1} \). Several oscillation periods are visible across this range, indicating complex Fermi surfaces5. To understand these data more quantitatively, we plot the Fourier transform of \( R_{xx}(1/B_i) \) as a function of \( f_c \), the oscillation frequency normalized to the total carrier density (Fig. 2b). \( f_c \) corresponds to the fraction of the total Fermi sea area enclosed by the Fermi surface generating the peak. Three regions of qualitatively different quantum oscillation spectra are visible. At extreme right, a single peak at \( f_c = 0.5 \) indicates two equal area Fermi surfaces each enclosing half of the total Fermi sea. We associate this regime with a spin-polarized, valley-unpolarized ‘half-metal’ state with a simply connected Fermi sea in each valley. At extreme left, several oscillation peaks with density-dependent frequencies are visible. These correspond to the inner and outer boundaries of an annular Fermi sea with the full fourfold spin and valley degeneracy (and harmonics). Intermediate between these two phases, the oscillation spectrum is more complex, including both strong peaks at \( f_c \leq 0.5 \) as well as at \( f_c \leq 0.1 \). We identify this regime with one or more partially isospin polarized (PIP) phases, where the system has broken one of the spin or valley symmetries but is not completely polarized into two isospin components. Comparing the quantum oscillation spectrum to base temperature transport measurements at \( B = 0 \) (Fig. 2c) shows that SCI occurs within the symmetric, annular phase and adjacent to the boundary with the PIP phase. The appearance of superconductivity so close to a symmetry-breaking phase transition opens the possibility of an unconventional superconducting state. A characteristic of many unconventional superconductors is their fragility with respect to disorder, due to the inapplicability of Anderson’s theorem23. Disorder in superconductors is quantified by the ratio of the coherence length at zero temperature (\( \xi_0 \)) to the perpendicular displacement field \( D \) measured with an 1 nA AC current at base temperature. \( f_c \), Temperature and current dependence of the differential resistivity \( dV/dI \) measured at the points in the \( n_e - D \) plane indicated in a. b, Temperature-dependent resistivity across SCI measured at \( D = 0.46 \text{ V nm}^{-1} \). c, \( R_{xx}(T) \) corresponding to the data plotted in f. \( T_{\text{FBT}} = 106 \text{ mK} \). Inset, device schematic showing measurement configuration \( R_{xx} \).
mean free path (\(\ell\)), \(d = \xi / \ell\), with the superconductivity destroyed when \(d = 1\) for unconventional superconductors. To assess \(d\) in RTG, we study the magnetoresistance of both the superconducting and normal states. Figure 3a, b shows the dependence of SC1 on the out-of-plane magnetic field \(B_{\perp}\). The critical \(B_{\perp}\) is in the 10 mT range. Within Ginzburg–Landau theory, \(B_{\perp}\) is related\(^2\) to the coherence length by \(2\pi \xi = \phi_0 / B_{\perp}\), where \(\phi_0\) is the superconducting flux quantum. As a result, \(\xi = 150–200\ nm\) for SC1, setting an upper bound for \(\xi\). As evident from the Drude conductivity \(\sigma = e^2 / m\ell\), where \(m\) is Planck’s constant, \(e\) is the elementary charge, \(k_F\) is the Fermi wave vector. Taking \(k_F = (1/2\pi m) = 0.25\ nm^{-1}\) and a normal state resistance of \(R = 20\ \Omega\) produces an estimate of \(d = 1\ \mu\) considerably larger than \(\xi\) and implying \(d \lesssim 0.2\). However, this estimate for \(d\) is comparable to the lateral dimensions of our device (Fig. 3c), calling into question the validity of the Drude approach. In fact, qualitative features suggest \(d\) may be considerably longer. Figure 3c, d shows a circuit schematic for measuring the nonlocal magnetoresistance, which has been used to detect transverse electron focusing in other graphene heterostructures. Measured data in the regime of SC1 (Fig. 3d) shows a pronounced feature near \(B_{\perp} = 0.1\ T\), consistent with transverse electron focusing between the contacts, which are separated by a pitch of \(L = 2.3\ \mu\). This feature, which is observed across all densities in our device (Extended Data Fig. 3), suggests \(L \approx 5\ m\ell / 2 = 3.5\ \mu m\). Taking this estimate for \(L\) gives a disorder parameter \(d \approx 0.05\). These estimates place the superconductivity firmly in the clean limit, where unconventional superconductivity may be expected to survive.

To further explore the properties of SC1, we show the response to an in-plane magnetic field in Fig. 3e. The in-plane critical field \(B_{\parallel}\) is several hundred milliTesla, more than one order of magnitude larger than \(B_{\perp}\), consistent with the two-dimensional (2D) nature of the superconductivity. To explore the mechanism for the magnetic-field-induced breakdown of superconductivity, Fig. 3f shows the dependence \(T_{\text{DKT}}\) on \(B_{\parallel}\). The data are well fit by the relation \(T_{\text{DKT}} = 1 - (B_{\parallel} / B_{\parallel}^{\text{crit}})^2\), for a superconductor limited by Pauli paramagnetism\(^2\). The extrapolated value of \(T_{\text{DKT}}\) is consistent with the Pauli temperature \(T_{\text{PK}}\), which is observed across all densities in our device (Extended Data Fig. 3).

**Fig. 4** | Fermiology and \(B_{\parallel}\)-dependence of SC2. a, Top, \(R_{xx}\) as a function of \(n_2\) and \(B_{\parallel}\) for \(D = 0.33\ \text{V nm}^{-1}\). Bottom, \(R_{xx}\) at \(B = 0\) for \(D = 0.33\ \text{V nm}^{-1}\). b, Fourier transforms of \(R_{xx}(B_{\parallel})\) for the values of \(n_2\) indicated by arrows in a. Insets, schematic Fermi contours. c, Temperature dependence of \(R_{xx}\) versus \(n_2\) measured at \(D = 0.33\ \text{V nm}^{-1}\). Temperatures between traces are equally spaced. d, Same as c, measured with an 0.99 T in-plane magnetic field applied. e, In-plane magnetic field dependence of \(R_{xx}\) versus \(n_2\) measured at \(D = 0.33\ \text{V nm}^{-1}\).

The critical \(B_{\parallel}\) is comparable to the lateral dimension \(l = 0.05\ \mu m\). These estimates place the superconducting gap \(\Delta\) in the clean limit, where unconventional superconductivity may be expected to survive.

To further explore the properties of SC1, we show the response to an in-plane magnetic field in Fig. 3e. The in-plane critical field \(B_{\parallel}\) is several hundred milliTesla, more than one order of magnitude larger than \(B_{\perp}\), consistent with the two-dimensional (2D) nature of the superconductivity. To explore the mechanism for the magnetic-field-induced breakdown of superconductivity, Fig. 3f shows the dependence \(T_{\text{DKT}}\) on \(B_{\parallel}\). The data are well fit by the relation \(T_{\text{DKT}} = 1 - (B_{\parallel} / B_{\parallel}^{\text{crit}})^2\), for a superconductor limited by Pauli paramagnetism\(^2\), where \(B_{\parallel}^{\text{crit}}\) and \(T_{\text{PK}}\) describe the \(T = 0\) critical field and \(B_{\parallel} = 0\) critical temperature, respectively.

We find \(\mu_B B_{\parallel}^{\text{crit}} / k_B T_{\text{PK}} = 1.7\), close to the value 1.23 predicted by weak coupling Bardeen–Cooper–Schrieffer (BCS) theory without accounting for the Coulomb repulsion or finite temperature effects. We thus conclude that the \(B_{\parallel}\) dependence is probably compatible with a conventional spin-singlet order parameter.

By contrast, the phenomenology of SC2 is not compatible with conventional spin-singlet pairing. As evident from the quantum oscillations shown in Fig. 4a, b, SC2 emerges from a twofold degenerate annular Fermi sea associated with a spin-polarized half-metal. Although the low \(T_{\text{C}}\) of SC2 complicates quantitative analysis of the kind presented for SC1, signatures of SC2 persist to very large values of \(B_{\parallel}\) with \(B_{\perp}\), and the critical current nearly unchanged for \(B_{\parallel}\) as large as 1 T (Fig. 4c–e and Extended Data Fig. 5). Taking a conservative estimate of 50 mK for \(T_{\text{C}}\) and 1 T for \(B_{\parallel}\), SC2 violates the Pauli limit of \(\mu_B B_{\parallel}^{\text{crit}} / k_B T_{\text{PK}} = 1.23\) (refs. \(^2\)) by more than one order of magnitude, consistent with a spin-polarized superconductor.

For attractive interactions of finite range, such as those that arise from electron phonon interactions, pairing potentials are attractive in all angular momentum channels. The potential is strongest in the s-wave channel, favouring conventional spin-singlet pairing in normal metals. In the spin-polarized half-metal regime where SC2 occurs, electrons with reversed spin are separated energetically from the ground state by the exchange energy, which is several meV (ref. \(^2\)), at least two orders of magnitude larger than observed superconducting gap implied by the low transition temperature. Spin-singlet pairing is thus energetically precluded. The unique properties of graphene nevertheless allow for superconductivity from conventional pairing mechanisms. Most importantly, the negligible spin-orbit coupling endows the spin-polarized half-metal with spinless time-reversal symmetry, which guarantees degeneracy between electrons in opposite valleys but with the same spin even in the absence of inversion.
symmetry. The smaller $T_c$ of SC2 relative to SC1 is consistent with pairing in a higher angular momentum channel by the same interaction. One natural order parameter, proposed for moiré systems with similar symmetries, is the spin-triplet, valley singlet ($c_{k+\mathbf{q}}^\dagger c_{-k}^{\uparrow}$) (refs. 29,30). This form of superconductivity shares many similarities with conventional superconductors, most notably protection from intra-valley scattering by smooth disorder potentials.

**Discussion**

The common features shared by SC1 and SC2 suggest several possible mechanisms, both conventional and all-electronic.

Most obviously, the appearance of superconductivity near symmetry-breaking phase transitions suggests that fluctuations of the proximal ordered state may have a role in pairing\(^{30}\). The plausibility of this picture hinges on the nature of the transition. Experimentally, the sudden jump in quantum oscillation spectra observed near the superconductors is suggestive of a first-order transition. In this case, fluctuations might be suppressed. However, the resistivity of the normal state changes only gradually across the transition, contrasting with other isospin transitions studied in the same sample that are strongly first order. Measurements of the thermodynamic compressibility similarly do not show strong negative compressibility where superconductivity is observed, allowing for the possibility of a continuous transition.

The nature of the proximal ordered state also has a key role in fluctuation-mediated superconductors, with different orders producing attraction in different pairing channels\(^{30}\). In RTG, in-plane field measurements show that the PIP phase proximal to SC1 is probably spin-unpolarized (Extended Data Fig. 7). To match the experiment, then, a theory of fluctuation-mediated superconductivity for SC1 should produce an apparently Pauli-limited superconductor from fluctuations of a spin-unpolarized isospin ordered state—a strong constraint.

Alternatively, superconductivity and symmetry breaking may arise in close proximity from unrelated mechanisms\(^{33,34}\). Within BCS theory, the superconducting transition temperature in the antiadiabatic limit\(^{33}\) applicable to low-density electron systems is approximated by

$$T_c = T_F e^{-1/\lambda}$$  \hspace{1cm} (I)

where the Fermi temperature $T_F = 50$ K in the regime of interest and $\lambda = g\lambda/s$ is the dimensionless coupling constant characterizing attractive interactions, which depends on the coupling constant $g$ and the density of states.

For a density-independent $g$ (as expected for phonon-mediated attraction, for example) superconductivity is observed at temperature $T$ when $\rho < \rho_{sc}$.

However, high density of states also favours symmetry breaking, with the boundary between ordered and disordered states defined by the Stoner criterion $\rho_{pm} > 1/U$ where $U$ parameterizes the Coulomb repulsion. As $\rho$ increases—a occurs in our experiment as $\rho_{sc}$ is reduced—one of two scenarios results. For $\rho_{pm} < \rho_{sc}$, the Stoner criterion is satisfied first, and the Fermi liquid becomes magnetic. As a result, $\rho$ decreases, the Kramers degeneracy is lifted, and superconductivity is not observed. Conversely, if $\rho_{pm} > \rho_{sc}$, superconductivity is observed. However, as the density of states is further increased above $\rho_{sat}$ the system nevertheless becomes magnetic. In this case, the domain of superconductivity is bounded from below by $\rho_{sc}$ and from above by $\rho_{pm}$. Superconductivity occurs at the cusp of a magnetic transition, precisely as observed, despite the lack of a causal link between the two.

Support for this scenario is bolstered by the fact that both superconductors arise at the threshold of a magnetic transition but are predominantly within the disordered phase; quantities such as $B_{c1}$ and $B_{c2}$ rise gradually as the isospin-symmetry-breaking transition is approached before rapidly collapsing at the transition itself. However, a key question that remains is whether this picture is consistent with the seemingly narrow range of $\rho_{sc}$ over which superconductivity is observed. For example, SC1 occurs over a density range $\Delta n/n = 5\%$. Comparing the maximum $T_c = 100$ mK to our estimated base temperature of 30–40 mK, we estimate $\Delta T_c/T_c = 0.6–0.7$ over this same range. For this to be accounted for entirely by a change in $\lambda$, $\Delta \lambda/\lambda = 0.1$, about twice as large as expected from single-particle calculations of the density of states. More detailed calculations (for example, accounting for both the Coulomb repulsion and finite temperature effects\(^{35}\)) may assess whether this quantitative discrepancy is important.

In both phonon- and fluctuation-mediated superconductors, high temperature transport typically shows signs of electron scattering by the same neutral modes that mediate the superconductivity\(^{36}\). We find no sign of enhanced high temperature scattering, at least up to 20 K (Extended Data Fig. 6). A mechanism for superconductivity (albeit not usually in the s-wave channel) that does not invoke soft modes was given in ref. 10 based on the intrinsic instability of the Fermi liquid. Although thought to occur only at experimentally inaccessible temperatures and disorder strengths in most materials, it has been proposed\(^{37}\) that in semiconductor quantum wells with two occupied subbands, this effect may be enhanced. Given the similarity between a two subband system and the annular Fermi seas we describe above, combined with the exceptionally low disorder in RTG, exploration of such mechanisms may be warranted.

In closing, we comment on the possible relationship between the superconductivity reported here and that observed in moiré systems. In RTG aligned to hexagonal boron nitride, the moiré potential only weakly perturbs the underlying isospin symmetry breaking\(^{38}\). The $\rho_{sc}$ and $B_{c2}$ dependence of the signatures of superconductivity observed in that system\(^{39}\) would appear to be most consistent with SC2. Twisted bilayer and twisted trilayer graphene have different microscopic symmetries; however, they share several features with RTG, including enhanced density of states and isospin symmetry breaking. We conjecture that the superconductivity observed in all graphene systems has the same underlying mechanism.

**Online content**

Any methods, additional references, Nature Research reporting summaries, source data, extended data, supplementary information, acknowledgements, peer review information; details of author contributions and competing interests; and statements of data and code availability are available at https://doi.org/10.1038/s41586-021-03926-0

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Methods

The trilayer graphene and hBN (hexagonal boron nitride) flakes were prepared by mechanical exfoliation of bulk crystals. The rhombohedral domains of trilayer graphene flakes were detected using a Horiba T64000 Raman spectrometer with a 488 nm mixed gas Ar/Kr ion laser beam. The rhombohedral domains were subsequently isolated using anodic oxidation cutting with an atomic force microscope37,38. The van der Waals heterostructures were fabricated following a dry transfer procedure26, with care taken to minimize the mechanical stretching of RTG. Fabrication details are described in ref. 7, which studied the same device.

Transport measurement was performed using a lock-in amplifier. Data in Extended Data Fig. 1 were measured at a frequency of 19.177 Hz. The rest data were measured at 42.5 Hz. The frequency was chosen to minimize electronic noise.

All measurements were performed in a dilution refrigerator equipped with a vector superconducting magnet. Unless specified, measurements were performed at base temperature, corresponding to $T \lesssim 20\,\text{mK}$ as measured by a calibrated ruthenium oxide thermometer mounted close to the sample. Cryogenic low-pass filters are applied to reduce the electron temperature.

Data availability

The data that support the findings of this study are available from the corresponding authors on reasonable request.

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Author contributions

H.Z. fabricated the device with assistance from T.X. H.Z. performed the measurements, advised by A.F.Y. K.W. and T.T. grew the hexagonal boron nitride crystals. H.Z. and A.F.Y. wrote the manuscript with input from all authors.

Competing interests

The authors declare no competing interests.

Additional information

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Extended Data Fig. 1 | Displacement field dependence of SC1. 

- **a**, $R_{xx}$ as a function of $n_e$ and $T$ at $D = 0.46$ V/nm.
- **b**, at $D = 0.4$ V/nm.
- **c**, at $D = 0.34$ V/nm.
Extended Data Fig. 2 | Temperature dependent data for SC1 and SC2.

a, Temperature dependent $dV/dI$ measurements of SC1. Measurements were performed at $n_s = -1.8 \times 10^{12} \text{cm}^{-2}$, $D = 0.46 \text{V/nm}$. b, $V(I)$ for SC1. The dashed line shows $V = I^3$; we take $T_{\text{BKT}}$ as the highest temperature where the $V(I)$ curve shows $I^3$ scaling. c, $R_T(T)$ for SC1 with $T_{\text{BKT}}$ indicated. d, Same as panel a, but for SC2. Measurements were performed at $n_s = -0.55 \times 10^{12} \text{cm}^{-2}$, $D = 0.33 \text{V/nm}$. e, Same as panel b, but for SC2. f, Same as panel c, but for SC2.
Extended Data Fig. 3 | Comparison of quantum oscillations and transverse magnetic electron focusing at $D = 0$. a, $R_{xx}$ vs $n_e$ and $B_\perp$ measured at $D = 0$. b, Fourier transform of $R_{xx}(1/B_\perp)$ for data in panel a. c, Non-local resistance measured in the configuration in Fig. 3c as a function of $n_e$ and $B_\perp$. 
Extended Data Fig. 4 | $B_z$ dependence of SC1. a, Temperature dependent $dV/dI$ measurements of SC1. Measurements were performed at $n_c = 1.8 \times 10^{12} \text{cm}^{-2}, D = 0.46 \text{V/nm}$. b, $R_{xx}(T)$ for SC1 with $T_{BKT}$ indicated. c, $V(I)$ for SC1. The dashed line shows $V \propto I^3$ scaling. d–f, Same as panel a–c measured at $B_z = 50 \text{mT}$. g–i, Same as panels a–c measured at $B_z = 100 \text{mT}$. j–l, Same as panels a–c measured at $B_z = 150 \text{mT}$. m–o, Same as panels a–c measured at $B_z = 175 \text{mT}$. where the $V(I)$ curve shows $I^3$ scaling.
Extended Data Fig. 5 | Magnetic field dependence of SC2. a. $R_{xx}$ vs $n_e$ measured at $D = 0.33$ W/nm and various $B_\perp$ with $B_\parallel = 0$. b. Same as a, measured at various $B_\parallel$ with $B_\perp = 0$. 
Extended Data Fig. 6 | Temperature dependence of $R_{xx}$ measured at $D = 0.4\text{V/nm}$ and $n_e < 0$. Bottom panel shows $R_{xx}$ as a function of $n_e$ at different temperature. Top panels show $R_{xx}$ vs $T$ at fixed $n_e$ extracted from the bottom panel.
Extended Data Fig. 7 | In-plane magnetic field dependence of the PIP phase near SC1. a, $B_y$ dependence of $R_{xx}$ near SC1 at $D=0.228\,V/nm$. b, Zoom-in of panel a. c, Same as panel b but measured with an out-of-plane field applied instead of in-plane field. d, Schematic phase diagram extracted from panel a. Insets are schematic Fermi contours of the isospin polarized and unpolarized phases.