Experimental results of SGS junctions employing W and Pt/Ta required to overcome the external noise. Although other with high sensitivity at low temperatures below 1 K are expected by the theory.22 Extensive studies on the electrical transport properties of our SGS junctions, which depend on the bias (V) and the back-gate voltage (V_{BG}), reveal the superconducting energy gap of 2Δ_{Pb} ∼ 2.2 meV. It is an order of magnitude higher than that of Al. The studies also reveal the subgap structures of differential conductance (dI/dV) induced by the multiple Andreev reflection.23 Thus, SGS junctions consisting of Pb_{1−x}In_{x} superconducting electrodes lead to the superior device performance over the previous Al-based SGS junctions.

II. SAMPLE PREPARATION

We used Pb_{1−x}In_{x} alloy instead of pure Pb as the superconducting-electrode material to minimize the granularity of electrodes while their T_{c} remains almost intact. Both lead (Pb) and indium (In) are type-I superconductors with different superconducting transition temperatures (T_{cPb} = 7.19 K, T_{cIn} = 3.40 K). The Pb_{1−x}In_{x} alloy, however, makes a type-II superconductor, T_{c} and critical magnetic field of which vary depending on the composition ratio.24 The Pb_{1−x}In_{x} film was thermally deposited at the rate of 0.7 nm/s in the base pressure of 6 × 10^{-7} Torr.

Atomic force microscope (AFM) measurement reveals that the Pb_{1−x}In_{x} (x = 0.04, 0.07, and 0.10) films exhibit a granular structure with the grain diameter of 300 ~ 400 nm and the root-mean-square roughness of 9.5 nm [see the inset of Fig. 1(a)]. It should be noted that the Pb_{1−x}In_{x} film on the graphene layer gets significantly thinner than the outer parts, which is probably caused by the fact that the Pb_{1−x}In_{x} is more mobile on graphene than directly on the oxidized Si substrate.25 T_{c} of

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**Observation of supercurrent in PbIn-graphene-PbIn Josephson junction**

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Superconductor-graphene-superconductor (SGS) junction provides a unique platform to study relativistic electrodynamics of Dirac fermions in graphene combined with proximity-induced superconductivity. We report the observation of the Josephson effect in proximity-coupled superconducting junctions of graphene in contact with Pb_{1−x}In_{x} (x = 0.07) electrodes for temperatures as high as T = 4.8 K, with a large value of I, R_{N} (∼255 μ V). This demonstrates that Pb_{1−x}In_{x} SGS junction would facilitate the development of the superconducting quantum information devices and superconductor-enhanced phase-coherent transport of graphene.

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I. INTRODUCTION

Properties of graphene, a monolayer honeycomb lattice of carbon atoms, have been investigated intensively ever since it was first discovered by the mechanical exfoliation.1,2 To date, experimental and theoretical studies have mainly been focused on features arising from its unique band structure, described by the relativistic Dirac equation.3–5 The chiral nature of charge carriers in graphene is revealed in transport measurements of the half-integer quantum Hall effect,6–8 weak localization,9,10 and Klein tunneling,11,12 etc.

A superconductor-graphene (SG) hybrid system, such as a superconductor-graphene-superconductor (SGS) junction or an SG interface, provides an ideal platform to investigate the relativistic nature of Dirac fermions combined with superconductivity.13,14 Instead of the retroreflection of carriers in an ordinary superconductor–normal-metal interface, an SG interface is theoretically predicted to show the specular reflection of chiral quasiparticle carriers.14 The specular reflection of carriers near the Dirac point can be more easily observed if superconducting material with a large superconducting energy gap is employed for the electrodes because the effect of unexpected charge doping near the Dirac point is minimized. Moreover, phase-coherent transport in graphene of quasiparticles with the energy below the superconducting energy gap can be enhanced significantly due to the condensation of electrons in superconductor into a single macroscopic quantum state. This property can be applied for mesoscopic phase interferometer15 of graphene. A large superconducting energy gap with a high transition temperature T_{c} also provides a potential advantage for qubit application of graphene, where a Josephson element is operable at high temperatures. Yet, previous experimental studies have mainly been focused on SGS junctions with Al electrodes16–19 which have a relatively small superconducting energy gap of 2Δ_{Al} ∼ 250 μ eV (this value is smaller than the bulk value of ∼340 μ eV) and low T_{c} (∼1 K). For Al-based SGS-junction devices measurements with high sensitivity at low temperatures below 1 K are required to overcome the external noise. Although other experimental results of SGS junctions employing W and Pt/Ta as superconducting electrodes have been reported,20,21 either the supercurrent was not clearly seen20 or it was achieved only after heavy annealing of the sample.21 Thus, realizing SGS Josephson coupling employing a new superconducting material with a higher superconducting energy gap, as in this study, can be regarded as significant progress in studies of superconducting-proximity effect in graphene.

We report on the fabrication and measurements of the SGS junctions employing Pb_{0.93}In_{0.07} as the superconducting-electrode material with a higher T_{c} of 7.0 K. The Pb_{1−x}In_{x}-based SGS junction exhibits the supercurrent up to T ∼ 4.8 K above the liquid-helium temperature, which is the highest operation temperature of an SGS junction reported to date. The junction response to the external magnetic or microwave field manifests the genuine Josephson characters expected by the theory.22 Extensive studies on the electrical transport properties of our SGS junctions, which depend on the bias (V) and the back-gate voltage (V_{BG}), reveal the superconducting energy gap of 2Δ_{Pb} ∼ 2.2 meV. It is an order of magnitude higher than that of Al. The studies also reveal the subgap structures of differential conductance (dI/dV) induced by the multiple Andreev reflection.23 Thus, SGS junctions consisting of Pb_{1−x}In_{x} superconducting electrodes lead to the superior device performance over the previous Al-based SGS junctions.
by an arrow. Left inset: topographic AFM image of the Pb0.93In0.07-based SGS junction in the normal state in a perpendicular field of $H = 4.2$ kOe and at $T = 6$ mK. The charge neutrality point ($V_{\text{CNP}} = -20$ V) is indicated by an arrow. Left inset: topographic AFM image of the Pb0.93In0.07 film showing granular morphology. Right inset: optical microscope image of the device. The dotted line denotes the boundary of the monolayer graphene. The narrow colored region indicates the 300-nm spacing of graphene layer between two Pb0.93In0.07 electrodes. The wide colored region of graphene layer used for the measurement of the quantum Hall plateaus, respectively. (b) Gate-voltage-dependent conductance of the narrow junction and the wide graphene sheet with the quantum Hall plateaus, respectively. (b) Gate-voltage-dependent conductance of the narrow junction and the wide graphene sheet with $H = 16$ T at $T = 6$ mK. The dotted line corresponds to $G = \nu e^2/h$ ($\nu = 2, 6, 10, \ldots$).

Pb$_{1-x}$In$_x$ film on graphene is sensitive to its thickness, which in turn is responsible for the superconducting proximity effect in graphene. The thickness and the atomic composition of Pb$_{1-x}$In$_x$ electrodes were adjusted so as to obtain the highest value of $T_c$ of 7.0 K for Pb$_{1-x}$In$_x$ film. The contact resistance obtained from three-probe measurement configuration was in the range of $1 - 15 \, \Omega \mu m^2$ in most of our devices and it was insensitive to the temperature change.

Monolayer graphene was mechanically exfoliated from the natural graphite by using Scotch brand tape and transferred on a highly electron-doped silicon substrate covered with a 300-nm-thick oxidized-silicon layer. The carrier density in the graphene was tuned by using the silicon substrate as a back gate. The 900-nm-wide electrode of Pb$_{0.93}$In$_{0.07}$/Au (200/10 nm in thickness) double layer was formed on top of a graphene flake with a spacing ($L$) of 300 nm between the electrodes. In this study, two sets of dilution fridges (Leiden Cryogenics Model MNK 126-500 and Oxford Instruments Model AST) were employed. The latter was used to measure the microwave response of the SGS junction. For the low-noise measurements, two-stage RC filters (cut-off frequency $\sim 30$ kHz) and $\pi$ filters were connected in series with the measurement leads and a low-frequency ($\sim 13.3$ Hz) conventional lock-in technique was adopted for the measurement of dynamic conductance $dI/dV$.

III. RESULTS AND DISCUSSION

Figure 1(a) shows the back-gate voltage ($V_{BG}$) dependence of the junction resistance at $T = 6$ mK, obtained by quenching the superconductivity of Pb$_{0.93}$In$_{0.07}$ electrodes in an external magnetic field of $H = 0.42$ T. It reveals the charge neutrality point ($V_{\text{CNP}}$) at $V_{BG} = -20$ V. The carrier mobility and the mean-free path are estimated to be $\mu = 1400$ cm$^2$/Vsec and $l = 24$ nm, respectively, for $\Delta V_{BG} = -30$ V, where $\Delta V_{BG} \equiv V_{BG} - V_{\text{CNP}}$. The spacing of the graphene junction between two Al electrodes is an order of magnitude longer than the carrier mean-free path ($L > l$) in the graphene, which implies that all the data should be analyzed in terms of the proximity-junction model in a diffusive limit.

The single-layered-ness of graphene is confirmed by the quantized Hall conductance measured in a high-magnetic field of $H = 16$ T. In a quantum-Hall regime, the conductance plateaus of $G = \nu e^2/h$ with the quantized filling factor of $\nu = 2, 6, 10$ are expected for the monolayer graphene, where $h$ is Planck's constant. However, the Hall conductivity ($\sigma_{xy}$) is mixed with the longitudinal conductivity ($\sigma_{xx}$), which is dependent on the aspect ratio ($\xi = L/W$) of the graphene layer in the junction area. Thus, conductance dips take place for the SGS junction with $\xi = 0.05$, while the neighboring junction with $\xi \sim 0.9$ on the same graphene layer exhibits the quantized conductance plateaus, as shown in Fig. 1(b).

The resistive transition of the Pb$_{0.93}$In$_{0.07}$ electrodes shows an onset of the superconductivity at $T_{c,\text{onset}} = 7.15$ K, while the electrodes become fully superconducting below $T_c = 7.0$ K [see the inset of Fig. 2(a)]. The Josephson coupling is established through the graphene layer in the SGS junction below $T_c$ of Pb$_{0.93}$In$_{0.07}$ electrodes. In Fig. 2(a), the current-voltage ($I$-$V$) curve obtained at the base temperature ($T = 6$ mK) shows a sharp switching from a dissipationless to a resistive state at a critical current ($I_c \sim 3.0$ $\mu$A). A reverse switching occurs at a retrapping current ($I_{R} \sim 0.8$ $\mu$A), which yields the clear hysteretic behavior. The ambient magnetic field...
was canceled by applying a compensation field of \( H = 1.84 \) Oe, at which \( I_c \) was maximized. Progressive evolution of the \( I-V \) characteristics with temperature is displayed in Fig. 2(b), where the bias current is swept from negative to positive polarity. The asymmetry in \( I-V \) curves represents the occurrence of hysteresis. With increasing \( T \), \( I_c \) becomes smaller and the hysteresis is reduced. It is noted that \( I_c \) remains finite for \( T \) as high as 3.83 K. After the thermal recycling (i.e., switching the cryostat from Leiden Cryogenics Model MNK 126-500 to Oxford Instruments Model AST), \( I_c \) became observable up to 4.8 K above the liquid-helium temperature (see the supplementary information).\(^25\) The corresponding critical current density, \( J_c(0) \sim 9.4 \times 10^{10} \) A/cm, is a record high obtained from SGS junctions to date.

Typical temperature dependencies of \( I_c \) and \( I_R \) are displayed in Fig. 2(c) for \( V_{BG} = -40 \) V. \( I_c \) decreases rapidly with increasing temperature but \( I_R \) remains almost constant up to \( T = 1.77 \) K, at which the hysteresis disappears (or \( I_c \) becomes equal to \( I_R \)). Similar \( T \) dependence of \( I_c \) is obtained for different gate voltages, as illustrated in Fig. 2(d). The overall \( I_c - T \) curves well fit the diffusive Josephson-junction behavior in a long junction regime,\(^{28}\)

\[
e(l, R_N = a E_{TH} \left[ 1 - b \exp\left( -a E_{TH} / 3.2 k_B T \right) \right])
\]

where \( E_{TH} \) is the Thouless energy and \( a \) and \( b \) are fitting parameters. From the \( R - V_{BG} \) curve in Fig. 1(a), \( E_{TH} = \hbar D / L^2 \); \( \hbar \equiv \hbar / 2 \pi \) is estimated to be about 90 \( \mu \)eV for \( V_{BG} \) sufficiently away from the CNP (\( \Delta V_{BG} = -40 \) V) and is weakly dependent on \( \Delta V_{BG} \). Here, \( D = (v_F l / 2); v_F \) is the Fermi velocity) is the diffusion constant of graphene. In comparison with the theoretical expectation of \( a = 10.8 \) and \( b = 1.30 \), in a long junction limit of \( E_{TH} / \Delta_{PbIn} \rightarrow 0 \), the best-fit values of parameters \( a \) and \( b \) of our device turn out to be 1.7 - 2.9 and \( \sim 1.3 \), respectively, with \( E_{TH} / \Delta_{PbIn} = 0.083 \). Our PbIn-based SGS junction, however, corresponds to an intermediate regime between the long-and short-junction limits (see the discussion below), where a smaller value of \( a \) is expected from the theoretical calculation of Ref. 28. This may lead to the reduction of the parameter \( a \) in our junction. We note that the onset temperature (\( T^* \)) of a finite \( I_c \) is strongly dependent on the gate voltage, resulting in the highly increased \( T^* \) and much larger \( I_c(T = 0) \) as well for the gate voltages further away from \( V_{CNP} \). This is attributed to a competition between the Josephson coupling energy (\( E_J = \hbar I_c / 2 \pi \)) and thermal fluctuations. Due to a small value of \( I_c \) near \( V_{CNP} \), the supercurrent is vulnerable to the thermal fluctuation, which results in the reduction of \( T^* \).

Gate-voltage dependencies of \( I_c \) and \( I_R \) at \( T = 6 \) mK are displayed in Fig. 3(a), the magnitude of which increases monotonously with \( \Delta V_{BG} \). As observed previously in SGS junctions,\(^15\) the \( V_{BG} \) dependence of \( I_c \) correlates with the \( V_{BG} \) dependence of the normal-state conductance \( G(V_{BG}) \) represented by the black curve. The significant difference between the two (\( I_c > I_R \)) indicates that the hysteretic \( I-V \) curves prevail over the whole \( V_{BG} \) even including \( V_{CNP} \). Similar hysteresis is observed in various proximity-coupled Josephson junctions consisting of normal metals,\(^{29}\) semiconductor nanowires\(^{30,31}\) and carbon nanotubes.\(^{32}\) In a resistively and capacitively shunted junction (RCSJ) model,\(^{22}\) which is a qualitative model for the Josephson junction, the hysteresis can be explained by the presence of a finite junction capacitance. Although the geometric capacitance (\( C \)) is supposed to be negligible in the SGS junction, the effective capacitance (\( C_{eff} = \hbar / R_N E_{TH} \)) can be suggested by replacing the quasiparticle characteristic time (\( \tau = R_N C \)) by the quasiparticle diffusion time in graphene between the electrodes (\( \tau_D = \hbar / E_{TH} \)),\(^{33}\) where \( R_N \) is the junction resistance in the normal state. Because of the gate-voltage independence of \( E_{TH} \), \( C_{eff} = 88 \) fF at \( \Delta V_{BG} = -40 \) V is inversely proportional to \( R_N \). The quality factor of the junction, which is defined as \( Q = (2e I_c R_N / E_{TH})^{1/2} \) in terms of \( C_{eff} \), varies in the range of \( Q \approx 1.8-2.3 \), depending on \( \Delta V_{BG} \). The approximate expression of \( Q \sim I_c / I_R \) yields a comparable result of 3.4–4.5, as can be estimated from Fig. 3(a). This supports our interpretation of the appearance of the hysteresis in terms of the effective capacitance \( C_{eff} \).

The \( I_c, R_N \) product, which is a figure of merit for a Josephson junction, is given in Fig. 3(b) as a function of \( \Delta V_{BG} \). The maximum \( I_c, R_N \) value reaches 255 \( \mu \)V at \( \Delta V_{BG} = -40 \) V and it reduces to 145 \( \mu \)V at \( V_{CNP} \). The ratio of the maximum \( I_c, R_N \) product to the superconducting gap energy turns out to be \( I_c R_N / \Delta_{PbIn} = 0.23 \), where the value of \( \Delta_{PbIn} = 1.1 \) meV determined by the multiple Andreev reflection (to be discussed below) is adopted. Combining with the ratio of \( E_{TH} / \Delta_{PbIn} = 0.083 \) obtained from the \( I_c - T \) curves in Fig. 2, the value of

\[
\text{FIG. 3. (Color online) (a) Gate-voltage dependence of } I_c \text{ (upper curve) and } I_R \text{ (lower curve) at } T = 6 \text{ mK with bias current swept from negative to positive polarity. (b) } I_c, R_N \text{ product as a function of the back-gate voltage. (c) } dI/dV \text{ vs } V \text{ curves with varying } \Delta V_{BG} = 0, -20, -40 \text{ V from bottom to top. The conductance with } \Delta V_{BG} = 0 \text{ is magnified by 2.5 times for the sake of clarity.} \]
\]
eIcRN/Δφn indicates that our PbIn-based SGS junction is in the intermediate regime between the short \((E_{TH}/Δφn > 1)\) and the long \((E_{TH}/Δφn < 0.01)\) junction limits. According to Ref. 28, the ratio of \(E_{TH}/Δφn\) is 0.083 of our device should correspond to \(eIcRN/Δφn < 0.5\) in the zero-temperature limit. The reduction of \(IcRN\) of our device below the expected value may be attributed to the suppression of \(Ic\) due to the thermal fluctuations and/or incomplete filtering of the external noise. It should be noted that the observed variation of \(IcRN\) with \(ΔV_{BG}\) can be attributed to \(V_{BG}\) dependence of \(E_{TH}\), except for the region of \(V_{BG}\) close to the CNP \((ΔV_{BG} < 10 \text{ V})\) where the carrier density strongly fluctuates due to the presence of electron-hole puddles.

When the highly transparent contact forms at the normal-metal–superconductor (NS) interface, the Andreev reflection process\textsuperscript{23,34} takes place, where an incident electron from the normal metal with energy below \(2Δφn\) is reflected as a hole and a Cooper pair propagates into the superconductor, or vice versa. In an SNS junction a quasiparticle in \(N\), accelerated away from \(V_{BG}\), is provided by a periodic modulation of \(Ic\). This multiple Andreev reflection (MAR) is responsible for the subgap conductance peaks occurring at voltages of \(Vn = 2Δφn/ne\), where \(n\) is an integer, as shown in Fig. 3(c). The corresponding superconducting energy gap of \(\text{Pb}_0.93\text{In}_{0.07}\) electrodes is estimated to be \(2Δφn = 2.2 \text{ meV}\), which is comparable to the bulk value of \(2.7 \text{ meV}\). The subgap structures are evident at the constant \(Vn\) values for different gate voltages and become clearer for \(V_{BG}\) away from \(V_{CNP}\). This is attributed to a longer phase coherence length of quasiparticles in a higher-doped region.\textsuperscript{35}

Direct evidence of the genuine Josephson coupling through the graphene layer, rather than any artifact such as an artifact such as an electron-hole puddle, is provided by a periodic modulation of \(dI/dV\) with \(f\) and \(\lambda\). In an SNS junction \(Ic\) and \(\lambda\) are well understood by the diffusive Josephson junction model. Temperature dependence of the dc and ac Josephson effects through the graphene layer are well understood by the diffusive Josephson junction model. The dc and ac Josephson effects through the graphene layer are sustained over the liquid helium temperature. Furthermore, the experimental play ground for the graphene-based Josephson

\[\Delta V_{BG}\] is the voltage interval between neighboring Shapiro steps. The voltage interval between neighboring Shapiro steps is obtained to be \(Vn = 12.4 \mu\text{V}\), which is irrespective of \(ΔV_{BG}\) and precisely corresponds to the theoretical expectation of \(Vn = hf/2e\). The proportionality relation between \(ΔVn\) and \(f\) with the slope of \(h/2e = 2.07 \mu\text{V/GHz}\) is also confirmed in the frequency range of \(f = 6–21 \text{ GHz}\) as shown in Figs. 4(c) and 4(d). The widths of the Shapiro steps in the current axis is quasiperiodically modulated with the microwave power \((P^{1/2})\), as shown in the inset of Fig. 4(a), which resembles the Bessel-function-like behavior.\textsuperscript{30} It is surprising to observe the quantized voltage plateaus at the elevated temperature up to \(T = 4.8 \text{ K}\) in Fig. 4(b), although the step edges are rounded by thermal fluctuations. The \(dI/dV\) curve in the inset indicate that the Shapiro steps are almost vanishing only at \(T = 5.4 \text{ K}\).

**IV. SUMMARY**

In summary, we demonstrate the realization of \(\text{Pb}_{1-x}\text{In}_{x}\)-based SGS Josephson junctions. Temperature dependence of the critical current and the value of \(IcRN\) product of our system are well understood by the diffusive Josephson junction model. The dc and ac Josephson effects through the graphene layer are sustained over the liquid helium temperature. Furthermore, the experimental play ground for the graphene-based Josephson
OBSERVATION OF SUPERCURRENT IN PbIn-GRAPHENE...