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Anomalous Coulomb drag between bilayer graphene and a GaAs electron gas

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Abstract

We report on Coulomb drag experiments between a bilayer graphene flake and a GaAs two-dimensional electron gas, where the charge-carrier densities of both systems can be tuned independently. For both p- and n-type graphene charge carriers, we observe that the Coulomb drag unexpectedly changes direction when the temperature is lowered. We find this phenomenon to be dominant when the Fermi wave vector in graphene is larger than in GaAs. At temperatures above ≈70 K, the drag signal is consistent with momentum exchange. In all discussed regimes, the Onsager relation is respected.

1. Introduction

Coulomb drag is a direct measure of the interactions between charge carriers located in two close-by conductors [1]. Moving charges in the ‘drive’ layer induce transport of charge carriers in the ‘drag’ layer via interlayer Coulomb interaction. In the study of two-dimensional electron gases (2DEGs), the quest towards the strongly interacting regime called exciton condensate [2, 3] raises the hope of realizing supercurrents, and thus dissipationless electronic devices [4].

Understanding the interactions between charge carriers in double-layer systems and reaching strongly interacting regimes has driven Coulomb drag research for more than a quarter century [1, 5]. Recently, graphene and two-dimensional materials have multiplied the possible material combinations for Coulomb drag experiments [6]. Progress in fabrications has led to the development of high-quality stacks of materials with small dielectric separations but negligible tunneling [7–9]. Furthermore, graphene has a different dispersion relation compared to most semiconductors and is ambipolar. It is thus a system of choice to study drag in a wide range of densities and with different kinds of charge carriers [10–13]. Still, many questions regarding the drag behavior in graphene systems remain open. For instance, a strong negative drag has been observed at low temperatures for double-bilayer graphene systems [12, 13] which has not been explained so far.

In this work, we have realized a combination of bilayer graphene with a GaAs 2DEG, allowing us to probe the interaction between charge carriers of the same or opposite polarities and with different dispersion relations. The first reported Coulomb drag experiment in a graphene/GaAs hybrid system has shown an increased drag resistance for low temperatures, which could be consistent with the onset of interlayer correlations [14]. Here, we present a system where the densities in both layers can be tuned separately, are independent of temperature, and where the interlayer leakage current is negligible. We restrict our study to measurement regimes where Onsager relations hold true. We show that Coulomb drag between bilayer graphene and a GaAs 2DEG exhibits two regimes. For \( T > 70 \) K, the drag signal is consistent with a momentum-exchange mechanism reaching the Fermi-liquid regime for high-enough densities. However, the drag reverses its direction for both charge-carrier types in graphene when the temperature and the GaAs 2DEG density are lowered. This result is similar to the behavior reported in a recent drag study between two bilayer graphene systems [12]. In our hybrid graphene/GaAs system, however, the reversed drag seems to depend on the Fermi wave vectors of both electronic systems.
2. Materials and methods

The different materials composing the hybrid systems are illustrated in the cross-sectional drawing in figure 1(a). A shallow GaAs/AlGaAs heterostructure was photolithographically patterned and contacted with annealed Au/Ge/Ni contacts. Ti/Au pads were defined by e-beam lithography (EBL) and evaporated on the surface (red in figure 1). Subsequently, a bilayer graphene flake was picked up with a large hexagonal boron nitride (hBN) flake using the Van der Waals stacking technique described in [8]. The number of graphene layers was verified using Raman spectroscopy [15, 16]. The graphene/hBN stack was then deposited onto the predefined contacts and additional Ti/Au top- and side-gates were patterned by EBL and evaporated (yellow in figure 1). An optical microscope image of the resulting device is shown in figure 1(b).

The GaAs/AlGaAs heterostructure has a doping layer located 16.4 nm below the surface. The measurements are all performed well below the temperature where the Si donors freeze out, which is expected around 150 K. No significant parallel conduction was detected at the highest temperature of measurement. Thus, we do not expect the doping layer to cause any screening between graphene and the 2DEG. The GaAs 2DEG is located 34 nm below the surface and has an average density over the entire mesa of \(5.1 \times 10^{11} \text{ cm}^{-2}\). When the temperature increases from 1.7 K to 100 K, this density varies by less than 10% and the mobility decreases from \(3 \times 10^7\) to \(1 \times 10^5 \text{ cm}^2\text{V}^{-1}\text{s}^{-1}\). Applying negative voltages to the two side gates \(SG_1\) and \(SG_2\) confines the underlying 2DEG into a channel. For sufficiently negative voltages, this channel is narrow enough to reside below the graphene flake only.

Interestingly, we note that the current flowing in the GaAs 2DEG shows a hysteretic behavior with respect to the side-gate voltages: this indicates the presence of charges at the interface between hBN and GaAs. These voltages were kept constant during the measurements to avoid the hysteretic regime. Three parameters were varied in the following measurements: the top gate voltage \(V_{\text{TG}}\), the interlayer voltage applied to graphene \(V_{\text{gr}}\), and the temperature \(T\). For all voltages applied and at all temperatures, the leakage current between graphene and the GaAs 2DEG was below 20 pA, which is small compared to the signal expressed in terms of current.

Figure 2 shows the characterization measurements of the 2D charge-carrier systems at 1.8 K. The GaAs 2DEG conductance in (a) gives the relative lever arm of both gate voltages \(d V_{\text{gr}}/d V_{\text{TG}}\) on the 2DEG (approximating the constant current line (white dots) by the green superimposed line and identifying it with a constant density condition in the 2DEG). The finite lever arm shows that the graphene density of states does not fully screen the electric field from the top gate.

The 2DEG magnetoconductance is plotted in figure 2(c), revealing Landau levels formed in the 2DEG under the graphene flake. Their known dependence on magnetic field and density leads to the graphene lever arm on the GaAs density \(d n_{\text{GaAs}}/d V_{\text{gr}}\). The obtained value is almost half as large as expected from the wafer’s nominal layer sequence. We will confirm this result with an electrostatic model. In figure 2(c), the GaAs 2DEG depletion occurs at \(V_{\text{gr}} = V_{\text{GaAs}} = -0.38 \text{ V}\). Knowing this point and the relative gate lever arm, one can determine the 2DEG depletion line, as plotted in red in figure 2(b).

The graphene 2-terminal conductance as a function of \(V_{\text{gr}}\) and \(V_{\text{TG}}\) is displayed in figure 2(b). The white dots locate the conductance minimum corresponding to the charge neutrality (CN) of the graphene flake residing on top of the GaAs heterostructure. The gate voltage dependence of the conductance dip is altered when the GaAs 2DEG is depleted. Indeed, when the 2DEG charged plane disappears from the graphene electrostatic environment, \(V_{\text{gr}}\) and \(V_{\text{TG}}\) become simply voltages applied to the two plates of a single capacitor. The slope of the graphene CN becomes close to 1 (compared with the white continuous line in figure 2(b)). The broad conductance dip gives a mobility \(\geq 5000 \text{ cm}^2\text{V}^{-1}\text{s}^{-1}\). This low value is probably due to its direct contact to GaAs.
Coulomb drag measurements

Coulomb drag is measured by applying a DC current ranging from $-1 \ \mu A$ to $+1 \ \mu A$ to the drive layer, which is either graphene or the GaAs 2DEG, and measuring the resulting DC voltage in the other system (the drag layer). Voltages are measured using a temperature-stabilized, low-noise DC amplifier [18]. For all measurements, the drive voltage (building up in the drive layer) is also monitored. The drive voltage is linear in drive current for all presented measurements and results in a maximum change in 2DEG density of $1 \times 10^{10} \ \text{cm}^{-2}$. The drag voltage (building up in the drag layer) is also linear in drive current above 30 K when the GaAs 2DEG drives. At lower temperatures, mesoscopic fluctuations appear in the graphene drag voltage [11, 19, 20]. The drag resistance is extracted from a linear fit of the drag voltage with respect to the drive current: this is the quantity plotted in figure 3. In this article, we take the sign convention that the drag resistance is the longitudinal transresistance from layer 1 to layer 2, $R_{\text{drag}} = R_{\text{2d}}^1/|S|$. When graphene drives, the GaAs drag voltage is linear with drive current above 70 K. A quadratic component appears below that temperature for low densities in graphene. This could be due to a Joule-like effect induced by the graphene-gold contact resistance and resulting in an energy transfer to the GaAs 2DEG. Onsager relations predict that exchanging the role of the two electronic systems should result in the same signal. This is quantitatively obeyed as shown in figure 3(a), when both drag voltages are linear, but
also for $T > 30$ K if one extracts the odd part of the drag signal in the 2DEG, which is linear. This means that the drag resistance can still be extracted from the GaAs drag signal below 70 K.

Figure 3 shows the drag resistance in the GaAs 2DEG (b) and in graphene (c) as a function of graphene and GaAs 2DEG densities at 100 K. A cut from these maps is shown in figure 3(a) for both configurations: as expected, the drag resistance is identical in both measurements except that the signal is noisier when graphene is used as the drag layer. This agrees with the observation that disorder in the drag layer yields noise in the drag signal [11].

We now show that the overall behavior of the high-temperature drag data agrees qualitatively with momentum-exchange-dominated Coulomb drag. Firstly, the drag decreases with increasing 2DEG density $n_{\text{Ga}}$ and increasing graphene density $n_{\text{gr}}$ away from charge neutrality. Secondly, the drag resistance between graphene holes and GaAs electrons is positive while, for electrons in both layers, it turns negative. This is explained by a picture where drag friction tends to equalize the charge-carrier velocities in both layers [1, 21]. The literature does not offer explicit calculations of the drag behavior between bilayer graphene and a GaAs 2DEG. However, a universal Fermi liquid regime following $R_{\text{drag}} \propto T^2/(n_1 \times n_2)^{3/2}$ should be reached in graphene, bilayer graphene, and 2DEGs when the charge-carrier densities and the interlayer separation are large enough [22–24]. For bilayer graphene and GaAs 2DEGs, this is achieved when $k_B T \ll E_F$ and $k_T d > 1$ [22, 25], with $d$ being the interlayer distance and $k_T$ the Thomas–Fermi screening wave vector at each layer, defined as $k_T = \frac{\varepsilon_s m_F^*}{4 e \varepsilon_r \varepsilon_0 h}$ [26]. In this formula, the degeneracy is called $g$, the effective mass $m^*$, and the dielectric constant between the two layers $\varepsilon_r$.

In our system, we find that $k_T d \approx 6 > 1$ and that the high-density or low-temperature regime is reached for $n > 2.5 \times 10^{11}$ cm$^{-2}$ in both the bilayer graphene and GaAs 2DEG. To check if our data fits the expected Fermi liquid behavior, we take cuts of the drag resistance at fixed density in one layer and plot it logarithmically against the density in the other layer in figure 4. The law is well obeyed for fixed 2DEG density, as seen in figures 4(a) and (b). It is also the case at fixed positive graphene density, although a small bump in drag resistance at $n_{\text{gr}} \approx 10 \times 10^{11}$ cm$^{-2}$ appears in figure 4(d). It may be due to the CN of graphene on gold, leading to a p–n junction at the contact border [12] (further explanations below). At negative $n_{\text{gr}}$, the drag data does not show a good agreement with the expected exponent (figure 4(c)). The reason for this discrepancy is unclear.

Close to the charge neutrality point, the drag resistance must switch sign, and the density dependence is far from the Fermi liquid regime. In a double-bilayer graphene system, the resulting peak is expected at around $E_F \approx a k_B T$, with $a \in [1, 2]$ depending on the flakes’ disorder [27]. In our hybrid system, the peak occurs for $n_{\text{gr}} \in [2.5; 3.2] \times 10^{11}$ cm$^{-2}$, which corresponds to 1.0 and 1.3 $k_B T$. This value compares well with the bilayer graphene calculations [27]. However, we will see that the drag temperature dependence exhibits a complicated behavior that does not allow for confirming this statement.

Finally, the isolevels of drag resistance in figures 3(b) and (c) feature sections which form almost straight lines, pointing towards a $n_{\text{gr}} + n_{\text{Ga}}$ dependence rather than the expected function of $n_{\text{gr}} \times n_{\text{Ga}}$. The same
observation has been reported for drag measurements in double-monolayer and bilayer graphene layers [11, 12]. However, the transition between the Coulomb drag behavior close to CN and the behavior at large densities should be considered. For instance, the isolevels of our device and of the ones just cited are very similar to the ones extractable from the computed Coulomb drag map for disorder-dominated monolayer graphene in figure 1 (right) of [28]. Thus, the observed isolevels might just be characteristic of the transition towards the Fermi-liquid regime.

Several more exotic effects should be considered before stating on a direct momentum transfer mechanism. The hydrodynamic regime reached at high temperature in ultra-clean graphene [29] is irrelevant in our low-mobility device. The energy-driven drag mechanism explained by Song and Levitov [30] can occur in the electron-hole puddle regime between two very close layers of graphene. We, on the contrary, have a weakly coupled drag system ($k_d > 1$) where only one drag partner can have an electron-hole puddle regime. Finally, phonon-assisted drag should lead to an enhanced drag signal along matched Fermi wave vector lines due to phase-space arguments [31]. This does not agree with our observations.

In summary, the high-temperature data measured in our hybrid device is consistent with the momentum-dominated mechanism of Coulomb drag due to (i) its charge-carrier-dependent sign, (ii) its power-law dependence on density, and (iii) the overall shape of the drag data, including a peak at positive and negative graphene density and isolevels in agreement with previous experimental [11, 12] and theoretical [28] works.

4. Anomalous Coulomb drag below 70 K

When the temperature is lowered, the drag resistance exhibits unexpected behavior. In figure 5, the drag voltage is measured in the GaAs 2DEG, graphene being the drive layer. Panel (c) shows the derivative of the graphene resistance (measured voltage across the drive layer divided by the applied drive current) with respect to the top-gate voltage. Like at 1.8 K (figure 2(b)), we see two peaks of resistance corresponding to the two aforementioned CN conditions in graphene. Figure 5(a) shows the drag resistance in the GaAs 2DEG as a function of $n_{gr}$ (adjusted via $V_{TG}$ ) and for different temperatures. The drag resistance as a function of both densities at 50 K is plotted in figure 5(b).

The CN of graphene on gold for $V_{TG} > -2$ V (panel (c)) leads to a Coulomb drag signal, including a sign change at low temperatures (panels (a) and (b)). This observation can be compared to the supplementary information of [12]: Li et al observed additional sign changes in the drag resistance between two bilayer graphene flakes when their top flake extended beyond the bottom one. These sign changes were due to a back-gate-induced doping in the top graphene outer leads, different from the central part of the top graphene. As a result, p–n junctions formed at the border of these leads, reduced the drive current, and disturbed the drag signal. In our system, the graphene parts above the gold contacts see an electric field of $(V_{TG} - V_{gr})/d_{BN}$ due to the top gate, with $d_{BN}$ the boron-nitride flake thickness. The main part of the graphene touching GaAs is subjected to the same electric field from the top gate but also an additional field due to the 2DEG. Hence a p–n junction can form at the interface between these two regions. The drive current remains constant despite this p–n junction because a change of a few kΩ in the graphene resistance is negligible compared to the 10 MΩ resistance in series with the drive current supply. One possible explanation for the drag signal at the graphene-on-gold CN could be that charge carriers are reflected from the p–n junction interface due to anti-Klein tunneling in bilayer graphene and disturb the drag. The temperature dependence of this excess drag resistance could be explained by the competition between thermal energy and the potential barrier of the p–n junction at the contact.

In the following, we focus on the region around the CN of graphene-on-GaAs, below $V_{TG} = -3$ V. The momentum-dominated drag picture breaks down as the temperature decreases. In figure 5(a), the drag-
resistance sign is reversed on both sides of the graphene CN as temperature is lowered from 100 to 33 K. Similar sign changes have been observed in systems of two graphene bilayer flakes [12, 13]. In Lee, Tutuc, et al’s work [13], a negative resistance peak appeared at the drag layer CN and was interpreted in terms of the energy transfer mechanism developed by Song and Levitov [30], which is valid for the electron hole puddle regime in graphene. In contrast, the reversed Coulomb drag observed in our work extends further in density than the puddle regime and changes sign at CN (see ‘R’ domains in figure 5(b)). Our observations are in better agreement with Li, Dean, et al’s results [12]. Indeed, the drag resistance is negative for the electron-hole regime and positive for the electron–electron regime for low 2DEG density. At higher 2DEG densities, the drag changes sign again to recover the expected signs for momentum–exchange-dominated drag (‘C’ domains). One can note that the anomalous drag has a larger magnitude than the conventional drag observed at higher temperatures (figure 3).

The two main differences with the previous observations [12] are the following: (i) in our sample, the drag resistance at low temperatures does not recover the expected sign at high absolute graphene density. This could be due to our limited range in that direction and to the spurious p–n junction effect appearing at high graphene electron density. (ii) The boundary between conventional and reversed sign at 50 K agrees with the condition of matched Fermi wave vectors in both 2D systems, marked by black dashes in figure 5(b). The agreement is qualitative and more satisfactory on the side of graphene holes than of electrons. However, this agreement is also visible if the roles of drag and drive layer are exchanged, i.e. Onsager relation is observed in this regime. It is also reproducible for different cool-downs. This sign change cannot be the result of the aforementioned p–n junction, because the charge carriers above the gold contacts change polarity far from the region of interest ($V_{\text{TG}} > -2$ V). This is in contrast to the report by Li et al, where the similar intrinsic doping inside the interaction area and in the leads did not allow for distinguishing between matched bilayer graphene densities and polarity switching in the leads (supplementary information of [12]). The derivative of the drive resistance with respect to top-gate voltage shown in figure 5(c) does not show any feature along the lines where the conventional drag sign is recovered. Thus, we can exclude p–n junction effects as a cause for the reversed drag.

The case of matched $k_F$ plays an important role in Coulomb drag because it corresponds to optimal momentum exchange between the layers. For instance, electron–hole pairing and exciton condensates are predicted to occur at matched Fermi wave vectors in strongly coupled double-layer systems. In our experiment, the drag resistance cancels out instead of increasing towards the single layers’ resistance, and thus the formation of electron–hole pairs is unlikely. Similarly phonon-enhanced drag that is optimized for matched $k_F$ [32] is also improbable. In hybrid graphene/GaAs systems, drag along the line of matched $k_F$ is expected to be independent of the interlayer distance, but only if the coupling is strong, which is not the case here [23, 24]. Our data suggests
that the relative importance of the mechanisms responsible for conventional and reversed drag could switch around the matched \( k_f \) condition, but the cause is not yet known.

The anomalous drag exhibits maxima along the graphene density axis, which agrees roughly with the estimated graphene disorder of around \( 3 \times 10^{11} \text{ cm}^{-2} \). This is in agreement with Li’s results and suggests that there is a relation between the reversed drag and graphene disorder.

Figure 5(a) introduces a further energy scale. Even though the sign change around CN is smooth, we can identify the temperature at which the transition occurs: it is close to 70 K, i.e. 6 meV. A plane capacitor estimate of the displacement field \( D \) across the bilayer graphene shows that the reversed drag occurs at \( D \lesssim 0.2 \text{ V/nm} \), corresponding to a band gap \( \lesssim 20 \text{ meV} \) [33]. However, hopping transport through localized states in the gap can significantly reduce the energy scale below which transport is suppressed [34]. Thus, the influence of the displacement field in this disordered device could disappear for temperatures above 70 K. Further experiments with less disordered graphene could help distinguish the different energy scales involved.

Since our findings are very similar to previous work on a double bilayer graphene system [12], the bilayer graphene band structure might be at the heart of the reversed drag. No drag reversal was reported in a system of two monolayer flakes [11]. However, preliminary measurements on a hybrid monolayer graphene/GaAs 2DEG device show indications of a sign change [17]. Further investigations in single-layer graphene hybrid devices are needed to reach conclusions as to an origin of the reversed drag lying in band structures.

Finally, hydrodynamic effects in the GaAs 2DEG’s channel under the graphene flakes could also be considered. Such effects should appear at low temperatures [35] and could alter the Coulomb drag behavior.

5. Conclusion and outlook

In summary, we report on Coulomb drag measurements in a hybrid system consisting of a bilayer graphene flake transferred onto a GaAs/AlGaAs heterostructure hosting a 34 nm deep 2DEG. Above 70 K, our observations point towards a conventional, momentum-exchange-dominated drag. When the charge-carrier density in one of the coupled two-dimensional systems is fixed, the conventional drag has a dependence on the other system’s density, in agreement with theoretical expectations for a weakly coupled, ballistic device in the Fermi liquid regime. The density-dependence isolevels of the drag resistance agree with those found in the Coulomb drag observed between graphene planes, both mono- and bilayers, at high temperatures [11, 12]. Their shape departs from the Fermi liquid regime behavior \( R_{\text{drag}} = f (n_{\text{G}} \times n_{\text{Ga}}) \). This could be due to the transition between the drag mechanism near the graphene charge neutrality and the high-density regime [28].

When the temperature is decreased below 70 K, a reverse drag appears. Our data agrees with a previous report on a double bilayer graphene system [12]. Further, the conventional drag seems to dominate the drag resistance for \( |k_f^D| < |k_f^{\text{C}}| \) whereas reversed drag dominates outside of this region.

Our experiments constitute the first study, to our knowledge, of Coulomb drag between bilayer graphene and a GaAs 2DEG where the density of each layer can be tuned separately. The interaction between graphene bilayer charge carriers and GaAs 2DEG electrons reveals a rich behavior that is not yet entirely elucidated. Reproducing these measurements between monolayer graphene and a GaAs 2DEG will help determining whether the bilayer graphene band structure is relevant for the anomalous drag.

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