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# Screen-printed and spray coated graphene-based RFID transponders

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#### Abstract

We report ultra-high-frequency (UHF, 800 MHz–1 GHz) radio frequency identification (RFID) transponders consisting of printed dipole antennas combined with RFID microchips. These are fabricated on Kapton via screen printing and on paper via spray coating, using inks obtained via microfluidization of graphite. We introduce a hybrid antenna structure, where an Al loop (small compared to the overall size of the antenna) is connected to a microchip with the double function of matching the impedances of antenna and microchip and avoiding bonding between exfoliated graphite and chip. The transponders have reading distance  $\sim 11$  m at UHF RFID frequencies, larger than previously reported for graphene-based RFID and comparable with commercial transponders based on metallic antennas.

#### 1. Introduction

Radio frequency identification (RFID) is a ubiquitous technology [1], with applications in access control [1], contactless payment [2], electronic passports [1], supply chain management [3], healthcare [4], food packaging [5] and animal identification [6]. It is also the cornerstone of the so-called 'Internet of Things' (IoT) [7], where people and devices are seamlessly integrated in a decentralized common set of resources, creating a convergence of the physical realm with human-made virtual environments [8]. Within IoT, every 'thing' is connected [8], and the already widespread RFID technology is likely to become even more ubiquitous, combining additional functionalities such as sensing [9,10] and energy harvesting [11,12].

The basic elements of a typical RFID system are tags [1] and readers [1], exchanging information via radio waves [1]. Tags comprise integrated circuits containing a memory to store the tag identity (ID) and the reading/writing circuitry [1]. Tags communicate with the reader via a suitable antenna, which typically has the double role of drawing energy from the reader to run the integrated circuit [1] and exchange data with the reader [1]. RFID offers advantages over other identification technologies, such as barcodes [1], since an RF tag does not need to be in sight of the reader and

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can, therefore, be embedded in objects [1]. Also, RFID allows simultaneous reading of several tags [1], making the identification process very fast, typically a few ms for passive (i.e. powered by the reader through the antenna) tags [1] and even shorter for active ones (i.e. battery powered) [1].

RFID tags should combine mechanical robustness (e.g. to tolerate vibrations) [1], light weight (typically <10g) [1], compact dimensions (~cm) [1], reliability [1] and low cost (<0.05\$) [13]. Mechanical flexibility might also be required (especially for IoT [14]), adding specific challenges not present on rigid systems, such as shifts in resonant frequency [14], and return loss (i.e. reflected power loss caused by antenna input impedance mismatch) [14] and changes in effective capacitance (i.e. the ratio of change in charge corresponding to change in potential) [14], radiation pattern distortion [14] and gain degradation [14]. Different operational scenarios also introduce additional complexity, e.g. proximity to tissues in wearables [14].

Large volume (several millions of units) [13] and low cost (<0.05\$ per unit) [13] manufacturability is essential, as it is expected that over one trillion IoT devices will be deployed by 2025 [15, 16]. The most common tags, consisting of a planar electric dipole antenna [17–19], are fabricated from a metallized plastic foil by acidic etching [1]. However, this process results in metal waste [20], which is also environmentally harmful [20].

Printing is a promising alternative [14], as it combines high volume production (e.g. an industrial screen printer can print areas >3 m × 6 m in a single pass [21]) and, at the same time, avoids chemical etching and material waste. Ag inks are typically used for printed RFID [22, 23], since they have high conductivity ~10<sup>6</sup> S m<sup>-1</sup> [24]. However, the Ag cost is very high (~800–1000\$ kg<sup>-1</sup>) [25]. Printed Ag films have limited flexibility, breaking at ~75% strain [26] and resistance increase up to ~15% upon bending [26].

Printed graphene layers can be an alternative to printed metals [27], as graphene combines good conductivity [27] and mechanical robustness [27]. Graphene can be dispersed in solvents (such as NMP [28] or water [28]), doped [28] or functionalized [28]. The surface resistivity of single layer graphene (SLG) at radio (300 KHz-300 MHz) and microwave (300 MHz-300 GHz) frequencies is higher than metals [29], resulting in losses [29] that prevent its use in antennas with high (>90%) efficiency (i.e. ratio between power irradiated by the antenna and power supplied) [29]. The SLG conductivity can be tuned by field effect [30], however, the changes are mostly in the real part [29], while in the imaginary part these are small, up to  $\sim 100$ GHz [31, 32], resulting in limited reconfigurability (i.e. tunability of radiation frequency, pattern or polarization) [33].

Thick (>1  $\mu$ m) exfoliated graphite films, consisting of few-layer graphene (FLG) flakes, can overcome such limitations, having sheet resistances  $R_S < 2 \ \Omega \ \Box^{-1}$  [34], corresponding to conductivities >10<sup>4</sup> S m<sup>-1</sup> [34]. These can also be deposited over large (m<sup>2</sup>) areas by screen printing or spray coating.

Screen printing is a common industrial technique for roll-to-roll patterned deposition [21]. Typical formulations of screen printable inks contain a conductive filler, such as Ag particles [26], and insulating additives (e.g. stabilizers and binders) [39], at a total concentration >100 g L<sup>-1</sup> [39]. Of this, >60 g l<sup>-1</sup> consists of the conductive filler needed to achieve sufficiently high (>10<sup>6</sup> S m<sup>-1</sup>) conductivities [26, 40]. Spray coating is also suitable for roll-to-roll production [41]. To the best of our knowledge, there are no reports on spray coated graphene-based antennas. However, spray coated FLG films with similar specifications to those needed for RFID antennas ( $R_S \sim 6 \Omega \square^{-1}$  and thickness ~8  $\mu$ m) were reported for use in electromagnetic interference (EMI) shielding [42].

A number of antennas based on solution-processed FLG films were reported [43–48]. Their reduced performance in gain and radiation efficiency compared to metallic antennas (typically over one order of magnitude [33]) is compensated by other functionalities, such as mechanical flexibility [48]. RFID transponders, based on FLG film antennas combined with RFID integrated circuits, were demonstrated [44, 45, 48], showing typical reading distance up to ~9 m [47]. This is smaller than commercial RFIDs, providing reading distances >10 m [44, 45, 48].

The input impedance of a typical RFID microchip at operating frequencies (865–868 MHz in Europe and 915 MHz in US [1]) is capacitive [1, 51], with a real part lower than the absolute value of the reactance [1, 51]. Thus, to match the impedance conjugately, i.e. to ensure that both microchip and antenna are electrically compatible with each other, the impedance of the antenna should be the complex conjugate to that of the microchip at the frequency of operation [22]. A two-branch dipole antenna might not have such a point on its impedance curve because of design [50], dimensions [50] or materials used [50]. The conjugate impedance match between microchip and antenna can be achieved by forming a loop inductor parallel to the feeding point on the antenna conductor [19].

Conductivity of printed graphene is lower than Al or Ag inks. Thus, this is taken into account in the design of the transponders, as they still have shorter reading distance than commercial ones.

Here, we present wideband RFID transponders with a hybrid Al-printed graphene antenna with reading distance competitive with commercial ones. These consist of graphitic antennas either screen printed on Kapton or sprayed on paper, coupled with a RFID chip through Al inductive loops, ensuring impedance matching, i.e. that the impedance of the antenna is the complex conjugate impedance of the microchip at the frequency of operation. The Al loop is significantly smaller than the overall antenna's size, therefore minimizing use of metal and not compromising the flexibility of the overall transponder. These have reading distances up to  $\sim 11$  m in the relevant UHF RFID bands: 865.6-867.6 MHz (Europe) and 902-928 MHz (USA and Japan), larger than graphene-based RFID tags previously reported [44, 45, 47, 48] and comparable with commercial RFID transponders [49].

#### 2. Antenna design

The antennas are designed using the electromagnetic simulation software high frequency structure simulator (HFSS) 15 (Ansys Inc. USA), assuming  $R_S \sim 3 \ \Omega \ \Box^{-1}$ , as typical for dried FLG films produced by microfluidizaton [34]. The two main parameters of a transponder antenna are input impedance [1], to match the antenna with the transponder microchip, and radiation efficiency, defined as the ratio of power radiated by the antenna and power supplied [50].

We use an Impinj Monza R6 UHF RFID microchip, with a 96 bits memory. This employs unregulated codes and is compatible with a wide range of tag form factors [51]. The input impedance is  $16 - j139\Omega$  at 915 MHz [51]. This is prevalently capacitive, with a real part lower than the absolute value of the reactance. Thus, to match the impedances conjugately, the antenna should have an impedance  $Z_{ant} = 16 + j139\Omega$  at the same frequency, i.e. it should be sufficiently inductive with







a low real part of the impedance. In order to achieve this, a parallel inductor in the dipole antenna is implemented as an opening on the conductor [44, 48].

We also introduce a hybrid structure in which we combine the printed FLG antenna with an Al inductive loop for impedance matching. The Al loop is significantly smaller than the overall size of the transponder, therefore minimizes the use of metals and does not compromise flexibility. The loop forms inductive coupling between microchip and antenna FLG conductor. Thus, no direct connection of microchip to FLG film is required.

We design and simulate FLG antennas using both FLG inductors and Al inductive loops. Both designs are made for the same FLG, with  $R_S \sim 3 \ \Omega \ \Box^{-1}$ . The optimized outer dimensions of the antenna, to work at 915 MHz with the FLG inductive loop, shown in figure 1, are 114 mm × 34 mm, and the dimensions of the opening are 13.3 mm × 10.1 mm. The outer dimensions of the hybrid antenna, figure 2, are the same. The dimensions of the upper opening of the antenna are 18.3 mm × 6 mm, and those of the lower opening are 18.3 mm × 20 mm.

The main tunable parameters, optimized by simulations, are the circumference of the loop and the length of the antenna. The first determines the input

Table 1. Dielectric n	naterials used	l in simu	lations
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	Thickness	ε <sub>r</sub> @915	$tan(\delta)@915$	
Material	$(\mu m)$	MHz	MHz	
PEN	125	2.6	0.01	This work
PET	50	2.8	0.01	This work
Fine paper	120	4	0.125	[52]

reactance [44, 48], while the latter determines the radiation resistance, i.e. the resistance caused by the radiation of electromagnetic waves from the antenna [53]. In the hybrid antenna, a rectangular opening is added, rather than a loop, to minimize Eddy currents induced by the inductive loop, since these would increase losses and decrease radiation efficiency. Shape and dimensions of the opening are chosen to minimize Eddy currents without significantly affecting antenna conductivity. The inductive loop, with  $14 \text{ mm} \times 6 \text{ mm}$  outer dimensions, is made of 0.8 mm wide and 9  $\mu$ m thick Al, figure 2. Simulations consider the FLG R<sub>S</sub> uniform, and include the dielectric substrates. The hybrid antenna is simulated using two substrates: 125  $\mu$ m thick polyethylene naphthalate (PEN) and 120  $\mu$ m thick fine paper. In the hybrid antenna, there is also a 50  $\mu$ m thick polyethylene naphthalate (PEN) as a carrier layer between

Transponder	Z <sub>ant</sub> (Ohms)	$L_Z(\mathrm{dB})$	$\eta$ (dB)	$D_{tag}$ (dB)	$R_{read}\left( m ight)$
Antenna with FLG inductive loop on PEN	77.5 + j138	-2.3	-5.4	3.2	8.9
Hybrid antenna on PEN	17.2 + j136	0	-4	3	13.1
Hybrid antenna on paper	17.9 + j137	0	-4.1	3	13

FLG and Al. The parameters of the dielectric layers are listed in table 1 and are measured using a HP 4291A impedance analyzer with a HP16453A dielectric material test fixture or are taken from reference [52]. The adhesive tape used to attach the Al loop into the antenna is not included in the final model as its effect on the antenna parameters is negligible. The antenna is not sensitive to the dielectric properties of the adhesive tape on the top of the loop. This is due to the operation of the loop as an inductor, in which the magnetic field dominates over the electric field.

Table 2 summarizes the simulated parameters at 915 MHz: input impedance  $Z_{ant}$ , attenuation due to impedance mismatch  $L_Z$ , radiation efficiency  $\eta$ , directivity (i.e. ratio between maximum radiation intensity in the main beam and average radiation intensity over all space)  $D_{tag}$  and calculated read range, i.e. the calculated maximum distance that the RFID tag can be read,  $R_{read}$ . As shown by table 2, the impedance of the antenna is not affected by the substrate materials, and the antenna dimensions remain the same between different substrates. The attenuation due to the impedance mismatch between antenna and microchip is calculated from the impedances as [50]:

$$L_Z = 1 - |(Z_{ant} - Z_{IC}^*)/(Z_{ant} + Z_{IC})|^2 \qquad (1)$$

where  $Z_{IC}$  is the complex impedance of the microchip. The forward-link (i.e. from reader to tag [54]) read range is calculated as [54,55]:

$$R_{read} = (c/4\pi f) \times (P_{txEIRP}D_{tag}\eta_{tag}L_Z/P_{ICsens})^{1/2}$$
(2)

where *c* is the speed of light, *f* is the frequency,  $P_{txEIRP}$  is the equivalent isotropically radiated power (i.e. measured radiated power in a single direction) of the reader device and  $P_{ICsens}$  is the read sensitivity of the microchip (i.e. minimum power required to activate the chip).  $P_{txEIRP} = 3.28$  W is the maximum allowed radiated power of a UHF RFID reader, as defined by the European regulatory environment for radio equipment and spectrum [56].  $P_{ICsens} = -20$  dBm, as specified for the Impinj Monza R6 microchip by the manufacturer [51]. Table 2 indicates that the transponder with a hybrid antenna has a longer read range (13.1 m). This is due to both better impedance match between antenna and microchip, and higher radiation efficiency.

#### 3. Experimental

Based on the design optimized by simulations, FLG antennas are fabricated either by screen printing or spray coating.

Inks are formulated by adding different amounts of rheology modifiers, after exfoliation of graphite, to tune the ink viscosity. Graphite flakes (Timrex KS25) are added to deionized (DI) water, at a concentration  $\sim 100 \text{ g L}^{-1}$ , and to sodium deoxycholate  $(\sim 5 \text{ g L}^{-1})$ . The mixture is processed using a microfluidizer (M-110P) at 207 MPa for 70 cycles. One cycle is defined as one pass of the liquid mixture through the interaction chamber, where high shear rate ( $\sim 10^8 \, \text{s}^{-1}$ ) is applied [34]. The exfoliated graphite flakes have a lateral size distribution peaked  $\sim 1 \ \mu m$  and thickness ~12 nm [34]. Microfluidization is a homogenization technique whereby high pressure (up to 207 MPa) is applied to a fluid [35], forcing it to pass through a microchannel (diameter <100  $\mu$ m). Other liquid phase exfoliation processes, such as sonication and shear-mixing, have low yields (<2% [36-38]) since shear forces are not applied uniformly [38]. The key advantage of microfluidization is that high shear is applied to the whole fluid volume [34], not just locally, subjecting all the material to intense shear forces.

Figure 3 plots a representative Raman spectrum, acquired by a Renishaw inVia at 514 nm excitation, of the processed material after microfluidization. The 2D peak consists of two components  $(2D_2, 2D_1)$ . Their intensity ratio changes from ~1.5 for the starting graphite to ~1.2, indicating exfoliation, but not complete to SLG [34, 57].

Following microfluidization, carboxymethylcellulose (CMC) sodium salt is added at a concentration  $\sim 10 \text{ g L}^{-1}$  to prepare a screen printable (SP) ink and  $\sim 5 \text{ g L}^{-1}$  for the spray coatable (SC) one. CMC acts as rheology modifier giving the SP-ink a viscosity ranging from  $\sim 570 \text{ mPa s at } 100 \text{ s}^{-1}$  to  $\sim 140 \text{ mPa s at}$  $1000 \text{ s}^{-1}$ , and to the SC one  $\sim 220 \text{ mPa s at } 100 \text{ s}^{-1}$  to 60 mPa s at  $1000 \text{ s}^{-1}$ .

The SP-ink is used to form FLG films both for antennas with FLG inductor and hybrid antennas on Kapton using a screen printer (Kippax KPX-2012) equipped with a 90 mesh per inch screen. These films are then annealed at 265 °C for 10 min to remove the binder and increase conductivity.  $R_S$  of the printed antennas measured using a four-point probe is ~5 $\Omega$   $\Box^{-1}$ , reduced to ~3 $\Omega$   $\Box^{-1}$  after annealing at 265 °C for 10 min. Figure 4(a) is a scanning electron microscope (SEM) image of a screen printed film after annealing. Annealing at higher temperatures or for longer times further reduces  $R_S$ , however it causes delamination from Kapton, making the antenna not usable.

The SC-ink is used for hybrid antennas and sprayed onto 3 substrates: (1) PEN, Q65HA-125  $\mu$ m; (2) multicoated matt art paper (Lumisilk-120  $\mu$ m);







**Figure 5.** Measurement setup of the transponder. The transponder under test (1) is taped on a piece of styrofoam (2) placed on a turntable (3). The measurement antenna (4) is connected to the Tagformance<sup>TM</sup> device. The distance between (1) and (4) is 500 mm.



(3) uncoated printing paper (Tesorp). The substrate is cut into the shape of the simulated antenna. SC is performed using a hand held manual spray pen for ~5 s, while moving over the antenna area, so that ink covers the whole substrate, resulting in a self-standing antenna. Air pressure is kept constant and the spraying distance is ~20 cm. The dry thickness of one pass is ~15–18  $\mu$ m. A SEM image of a FLG film on paper is in figure 4(b).

The uncoated paper absorbs the water from the ink and the samples are dried and flattened using a hot press at  $\sim$ 130 °C. The samples are then calendered using a cylinder press with one steel and one hard rubber roller, generating a pressure  $\sim 80$  bar ( $\sim 36$  kN  $m^{-1}$ ). The compression is performed at 2 m min<sup>-1</sup> and up to 3 times. The adhesion of the dry ink on plastic and multicoated paper is not optimal, so this process is only done for uncoated printing paper, where the ink is more easily absorbed deep into the substrate.  $R_S$ is measured by four-probe close to the centre of the antenna, where the highest conductivity is required, as shown in the simulations in figures 1 and 2. R<sub>S</sub> saturates at  $\sim$ 3.6  $\Omega$   $\Box^{-1}$  after 2 spray passes. Further calendaring or additional coating do not reduce R<sub>s</sub>. The reason is that paper fibres limit the conducting pathways

available for the FLG flakes, as the ink is absorbed into the substrate before it can dry, due to the FLG concentration and the evaporation of water. SEM images of SP films on Kapton and SC on paper are shown in figures 4(a) and (b).

For the transponder with FLG inductor, the microchip is glued directly to the antenna using Ag paste. For the hybrid system, the Al inductive loop is fabricated similarly to conventional dipole transponders [1], i.e. by etching Al on PET [1]. The microchip is subsequently attached onto the Al loop using anisotropic conducting adhesive (ACA) [58] and the loop is attached on the antenna with adhesive tape.

The benefits of using a separate metal loop with an inductive coupling to the antenna radiator are based on using high conductivity metal (such as Al or Cu, with  $R_S \sim 3 \text{ m } \Omega \square^{-1}$ ) as the loop material (as the current density is highest in the loop), which makes the loop conductor narrow (~0.8 mm), and the loop small (~14 mm × 6 mm). The microchip is then easy to attach with existing industrial processes [59, 60]. Due to low loss in the metal loop, the efficiency of the antenna is higher. The loop is significantly smaller than the overall antenna size, minimizing use of metal, not compromising the flexibility of the transponder.





Figure 8. Measured (solid line) and simulated (dashed line) directivity of antenna with FLG inductive loop on PET in azimuth and elevation plane.



Figure 9. Measured (solid line) and simulated (dashed line) directivity of hybrid antenna on PET in azimuth and elevation plane.

Using an Al inductor loop, not only improves impedance matching in terms of conjugate impedance, but also reduces signal attenuation between antenna and microchip. Indeed, forming a contact between FLG antennas and microchip is challenging, especially considering the small ( $\sim$ 400  $\mu$ m  $\times$  250  $\mu$ m) contact pads of an RFID microchip. ACA, typically used with metallic tags [58], does not necessarily work on FLG, due to the temperature and pressure required by the bonding process [61]. Therefore, similar to reference [62], for the antenna with FLG inductive loop we use Ag paint to establish an electrical contact between FLG films and RFID chip, figure 1(a). Conversely, in our hybrid design, the printed FLG antenna and the RFID chip are connected through the Al loop and no bonding or Ag paint is required between loop and FLG antenna. Therefore, conventional ACA can be used to bond the RFID chip to the Al loop.

#### 4. Results and discussions

Figure 1 shows image and simulated current distribution of the antenna with parallel inductor, implemented as an opening on the FLG conductor. The current is concentrated around the opening or the loop inductor of the transponder, figure 1(b). The hybrid antenna is shown in figure 2(a). Figure 2(b) is the corresponding simulated current distribution. The highest density of current is in the metal conductor, thus maximizing power transfer to the microchip, therefore improving the reading range.

The measurement setup of the transponder is shown in figure 5. The antennas are measured with a Tagformance<sup>TM</sup> UHF RFID measurement system [63] in its anechoic cabinet. The evaluation is based on measuring the activation level of the transponder in a fixed and known setup [54, 64]. The transponders are attached on a piece of Styrofoam, acting as radiationtransparent support. The measured activation level is then used to calculate the theoretical reading range (i.e. the maximum range) in figures 6 and 7. The simulated reading ranges are also included for comparison.

For all antennas, the measured read range is shorter than simulations. However, for the hybrid antenna the discrepancy is smaller. A possible cause for this is the roughness of the edges in the SP antennas. Figure 1 indicates that the current concentrates on the edges of the opening or the loop in the middle. Thus, any added resistivity there has a significant impact on losses. This also explains why the difference between simulations and measurements is greater for antennas with the FLG inductive loop. These also use Ag paste as the conductor between antenna and microchip. The connections between Ag paste and FLG, as well as between Ag paste and microchip contact pads, are likely to introduce additional contact resistance, hence signal attenuation. The contact resistance between microchip pads and FLG, and the roughness of the loop inner edge, are the main reasons for the difference between simulations and experiments. The impact of the contact resistance was studied in reference [65]. The roughness of the loop inner edge is apparent by inspection using a microscope, but it is difficult to model electromagnetically. As the RF current concentrates on this inner loop edge, the effect on losses may be large.

Figure 7 shows that the reading range of SP and SC antennas are almost identical. Only below  $\sim$ 880 MHz the distance of SP antennas is  $\sim$ 10% smaller than SC,

showing how both deposition methods are suitable for the realization of FLG antennas.

The radiation patterns are also measured with the Tagformance<sup>TM</sup> system. Figures 8 and 9 compare measured directivities (solid red lines) and simulations (dashed blue lines). As the absolute directivity is difficult to measure, the measured radiation patters are normalized to the simulated ones at  $\phi = 0$ ,  $\theta = 0$ .

Radiation patterns, both simulated and measured, reveal a small difference compared to an ideal dipole antenna. The radiation pattern is not perfectly round on the azimuth plane. The difference in directivity between 0 and 180° is 2.8 dB for the FLG inductive loop antenna and 1.7 dB for the hybrid one. This can also be seen on the elevation plane. The maximum directivity in table 1 is above the theoretical one of a dipole antenna ~2.15 dBi (decibels relative to isotropic radiator) [50]. This can be attributed to the asymmetry of the transponders combined with the FLG  $R_S$ .

#### 5. Conclusions

UHF RFID transponders with screen-printed and sprayed FLG antennas were designed, fabricated and tested. Read ranges ~6.7 and 11.1 m were measured for antennas with an FLG inductive loop and for hybrid antennas, respectively. The transponders operate at the frequency bands reserved for UHF RFID: 865.6–867.6 MHz (Europe) and 902–928 MHz (USA, Japan). The hybrid antenna has reading performance superior to previously reported graphene-based RFID tags [44, 45, 47, 48] and comparable with commercial ones [49]. It also avoids the need for a direct contact between FLG film and microchip, making the fabrication of FLG antennas compatible with existing industrial processes.

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