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To cite this article before publication: Kaarle Jaakkola *et al* 2019 *2D Mater.* in press <https://doi.org/10.1088/2053-1583/ab48d8>

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Screen-printed and spray coated graphene-based RFID transponders

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We report Ultra-High-Frequency (UHF, 800MHz-1GHz) 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~11m at UHF RFID frequencies, larger than previously reported for graphene-based RFID and comparable with commercial transponders based on metallic antennas.

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 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 to corresponding 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 > 3m×6m in a single pass[21]) and, at the same time, avoids chemical etching and material wastage. Ag inks are typically used for printed RFID[22, 23], since they have high conductivity~10⁶S/m[24]. However, the Ag cost is very high (~800-1000\$/kg)[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 (300KHz to 300MHz) and microwave (300MHz to 300GHz) 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~100GHz[31, 32], resulting in limited reconfigurability (i.e. tunability of radiation frequency, pattern or polarization)[33].

Thick (> 1μm) exfoliated graphite films, consisting of

few-layer graphene (FLG) flakes, can overcome such limitations, showing sheet resistances $R_S < 2\Omega/\square$ [34], corresponding to conductivities $>10^4\text{S/m}$ [34]. These can also be deposited over large (m^2) 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 inks contain a conductive filler, such as Ag particles[26], and insulating additives (e.g. stabilizers and binders)[39], at a total concentration $>100\text{g/L}$ [39]. Of this, $>60\text{g/L}$ consists of the conductive filler needed to achieve sufficiently high ($> 10^6\text{S/m}$) 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 ($R_S \sim 6\Omega/\square$ and thickness $\sim 8\mu\text{m}$) 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 $\sim 9\text{m}$ [47]. This is smaller than commercial RFIDs, providing $>10\text{m}$ [44, 45, 48].

The input impedance of a typical RFID microchip at operating frequencies (865–868 MHz in Europe and 915MHz 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, even if the lower conductivity is taken into account in the design of the transponders, they still have shorter reading distance than commercial ones.

Here, we present a wideband RFID transponder with a hybrid Al-printed graphene antenna with reading distance competitive with commercial ones. Our transponders 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 antennas size, therefore minimizing use of metal and not compromising the flexibility of the overall transponder. These have reading distances up to $\sim 11\text{m}$ in the relevant UHF RFID bands: 865.6–867.6MHz (Europe) and 902–928 MHz (USA and Japan), larger than graphene-based RFID tag previously reported[44, 45, 47, 48] and comparable with commercial RFID transponders[49].

ANTENNA DESIGN

The antennas are designed using the electromagnetic simulation software High Frequency Structure Simulator (HFSS) 15 (Ansys Inc. USA), assuming $R_S \sim 3\Omega/\square$, as typical for dried FLG films produced by microfluidization[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 915MHz[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 a 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 minimizing 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 $R_S \sim 3\Omega/\square$. The optimized outer dimensions of the antenna to work at 915 MHz with the FLG inductive loop, shown in Fig. 1, are $114\text{mm} \times 34\text{mm}$ and the dimensions of the opening are $13.3\text{mm} \times 10.1\text{mm}$. The outer dimensions of the hybrid antenna, Fig.2, are the same. The dimensions of the upper opening of the antenna are $18.3\text{mm} \times 6\text{mm}$, and those of the lower opening are $18.3\text{mm} \times 20\text{mm}$.

The main tunable parameters of the antennas, optimized by simulations, are the circumference of the loop and the length of the antenna. The first determines the input reactance of the antenna[44, 48], while the latter determines the radiation resistance, i.e. the resis-

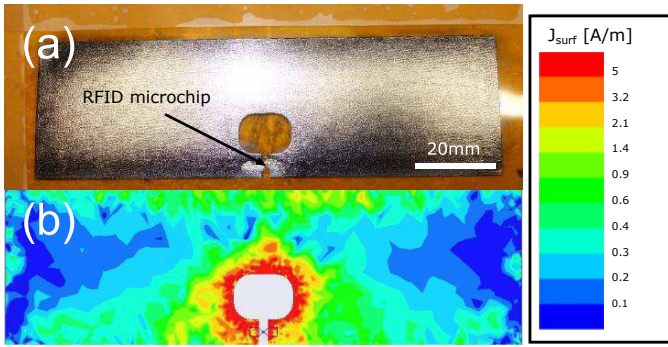


FIG. 1. a) Antenna with FLG inductor. b) Simulated current distribution. J_{surf} is the surface current density in A/m

Material	thickness (μm)	ϵ_r @915MHz	$\tan(\delta)$ @915MHz	
PEN	125	2.6	0.01	this work
PET	50	2.8	0.01	this work
fine paper	120	4	0.125	[52]

TABLE I. Dielectric materials used in simulations

tance 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 0.8mm wide and $9\mu\text{m}$ thick Al, Fig.2. Simulations consider the FLG R_S as uniform. The dielectric substrates are included in the simulation. Under the FLG antenna, there is a layer of dielectric. The hybrid antenna is simulated using two substrates: $125\mu\text{m}$ thick PEN and $120\mu\text{m}$ thick fine paper. In the hybrid antenna, there is also a $50\mu\text{m}$ thick PET as a carrier layer between FLG and Al. The parameters of the dielectric layers are listed in Table I and are obtained by measurement using a HP 4291A impedance analyzer with a HP16453A dielectric material test fixture or from Ref.[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 II summarizes the simulated parameters at 915MHz: input impedance Z_{ant} , attenuation due to impedance mismatch L_Z , radiation efficiency η , directivity (i.e. ratio between maximum radiation intensity in the main beam and average radiation intensity over

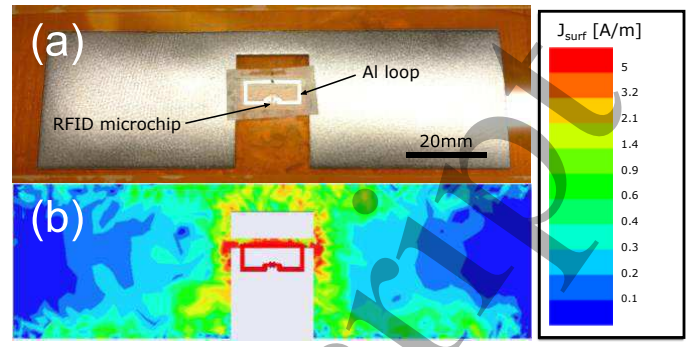


FIG. 2. (a) Hybrid antenna with metal inductor. The larger structure is the printed FLG, while the inner loop is the Al inductor. b) Simulated current distribution. J_{surf} is the surface current density in A/m.

Transponder	Z_{ant} (Ohms)	L_Z (dB)	η (dB)	D_{tag} (dB)	R_{read} (m)
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

TABLE II. Simulated parameters of the tag antennas at 915MHz

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 II, 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]:

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

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]:

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

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). $P_{txEIRP}=3.28\text{W}$ is the maximum allowed radiated power of a UHF RFID reader as defined by the European regulatory environment for radio equipment and spectrum[56]. $P_{ICsens}=-20\text{dBm}$, as specified for the Impinj Monza R6 microchip by the manufacturer[51]. Table II indicates that the transponder with a hybrid antenna

has a longer read range (13.1m). This is due to both better impedance match between antenna and microchip, and higher radiation efficiency.

EXPERIMENTAL

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

Inks suitable for screen printing and spray coating 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 $\sim 100\text{g/L}$ and sodium deoxycholate ($\sim 5\text{g/L}$). The mixture is processed using a microfluidizer (M-110P) at 207MPa 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 at $\sim 1\mu\text{m}$ and thickness $\sim 12\text{nm}$ [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 $< 100\mu\text{m}$). 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.

Fig.3 plots a representative Raman spectrum, acquired by a Renishaw inVia at 514nm 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 $\sim 10\text{g/L}$ to prepare a screen printable (SP) ink and $\sim 5\text{g/L}$ 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 100s^{-1} to $\sim 140\text{mPa s}$ at 1000s^{-1} , and to the SC one $\sim 220\text{mPa s}$ at 100s^{-1} to 60mPa s at 1000s^{-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 are then annealed at 265°C for 10 minutes to remove the binder and increase conductivity. R_S of the printed antennas measured using a four-point probe is $\sim 5\Omega/\square$, reduced to $\sim 3\Omega/\square$ after annealing at 265°C for 10mins. Fig.4a is a scanning electron microscope (SEM) image of the 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.

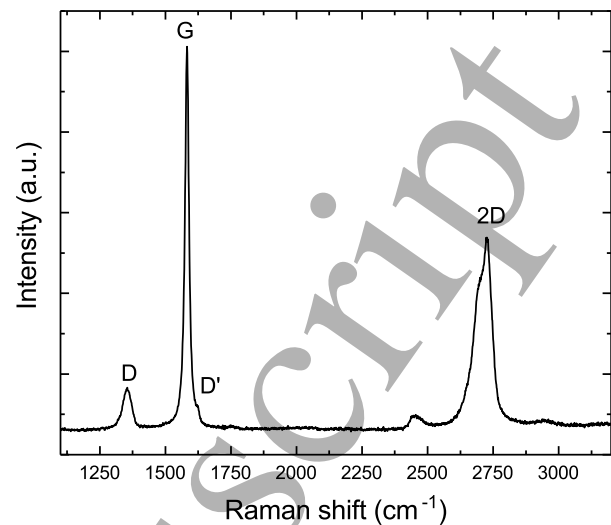


FIG. 3. Representative Raman spectrum at 514nm for flakes processed for 70 cycles.

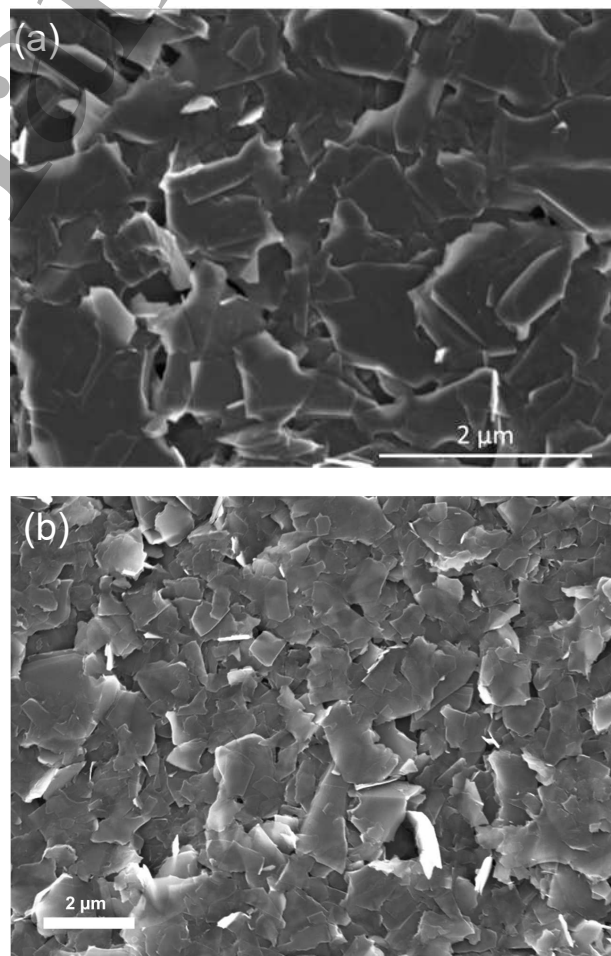


FIG. 4. SEM images of a) SP film on Kapton; b) SC film on paper

The SC-ink is used for hybrid antennas and sprayed onto 3 substrates: 1) Polyethylene Naphthalate (PEN), Q65HA-125 μm ; 2) multicoated matt art paper (Lumisilk-120 μm); 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 $\sim 5\text{s}$, 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 $\sim 20\text{cm}$. The dry thickness of one pass is $\sim 15\text{--}18\mu\text{m}$. A SEM image of a FLG film on paper is in Fig.4b.

The uncoated paper completely absorbs the water from the ink and the samples are dried and flattened using a hot press at $\sim 130^\circ\text{C}$. The samples are then calendared using a cylinder press with one steel roller and one hard rubber roller, generating a pressure $\sim 80\text{bar}$ ($\sim 36\text{kN/m}$). The compression is performed at 2m/min 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 Figs.1,2. R_S saturates at $\sim 3.6\Omega/\square$ after 2 spray passes. Further calendaring or additional coating do not further reduce R_S . 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 Figs.4a,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 polyethylene terephthalate (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 $R_S \sim 3\text{m}\Omega/\square$) as the loop material (as the current density is highest in the loop), which makes the loop conductor narrow ($\sim 0.8\text{mm}$), and the loop small ($\sim 14\text{mm} \times 6\text{mm}$). 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. 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 ($\sim 400\mu\text{m} \times 250\mu\text{m}$) contact

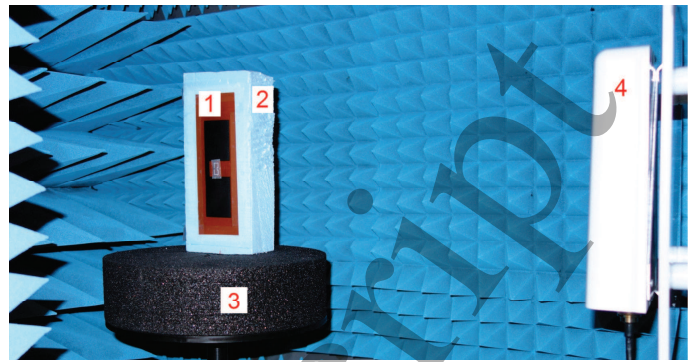


FIG. 5. Measurement setup of the transponder. The transponder under test (1) is taped on a piece of styrofoam (2) that is placed on a turntable (3). The measurement antenna (4) is connected to the tagformance device. The distance between (1) and (4) is 500mm

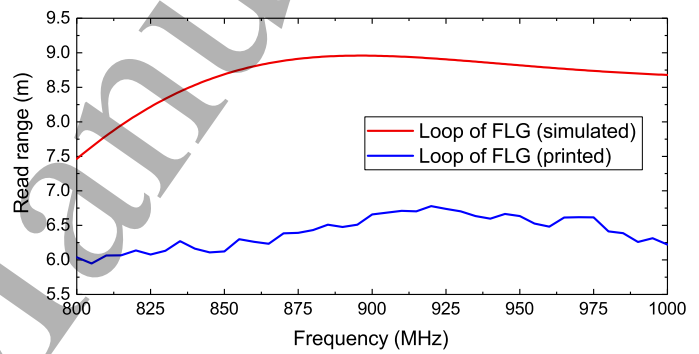


FIG. 6. Simulated and measured read range as a function of frequency for antenna with FLG inductive loop on PET

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 Ref.[62], for the antenna with FLG inductive loop we use Ag paint to establish an electrical contact between FLG films and RFID chip, Fig.1a. Conversely, in our hybrid design, the printed FLG antenna and the RFID chip are con-

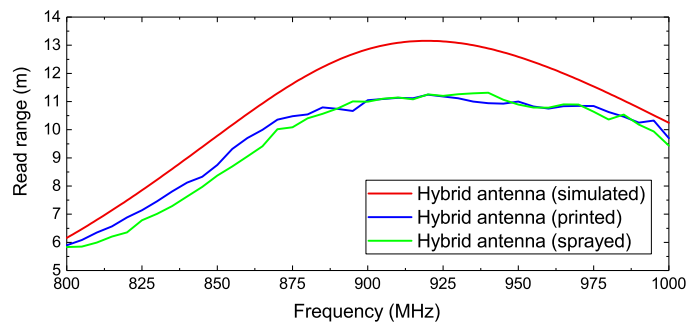


FIG. 7. Simulated and measured read range as a function of frequency for hybrid antenna on PET

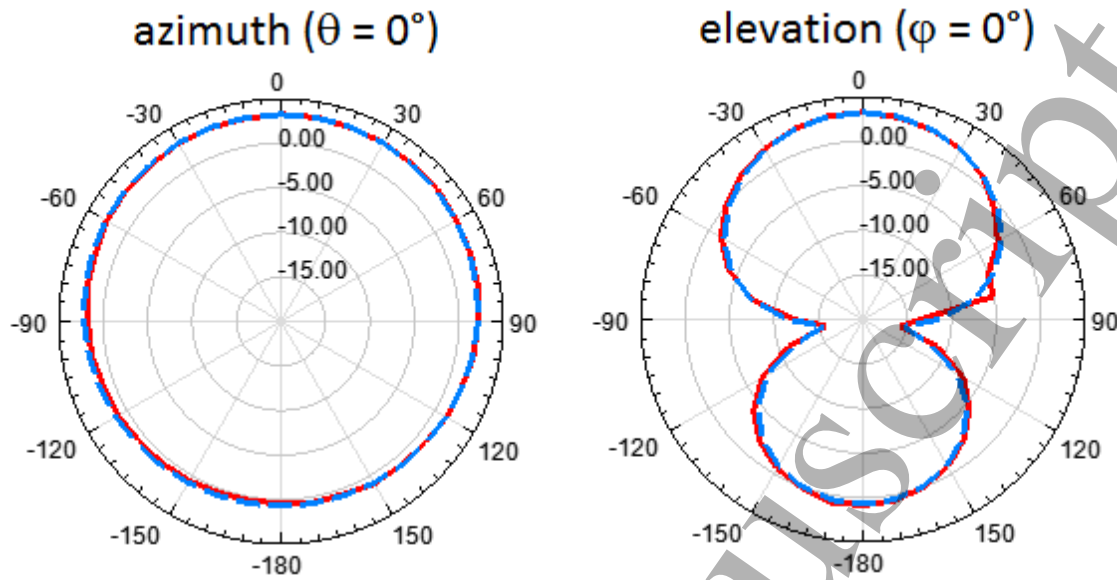


FIG. 8. Measured (solid line) and simulated (dashed line) directivity of antenna with FLG inductive loop on PET in azimuth and elevation plane

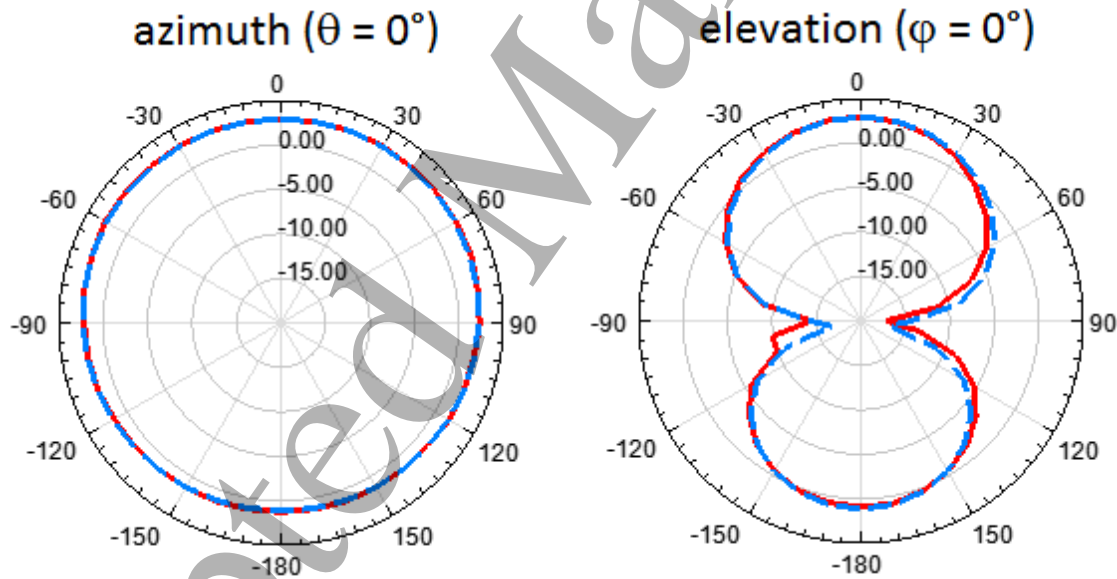


FIG. 9. Measured (solid line) and simulated (dashed line) directivity of hybrid antenna on PET in azimuth and elevation plane

nected 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.

RESULTS AND DISCUSSIONS

Fig.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, Fig.1b. The hybrid antenna is shown in Fig.2a. Fig.2b 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 Fig.5. The antennas are measured with a TagformanceTM 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 radiation-transparent support. The measured activation level is then used to calculate the theoretical reading range (i.e. the maximum range) in Figs.6,7. 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. Fig.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 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 the 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 Ref.[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.

Fig.7 shows that the reading range of SP and SC antennas are almost identical. Only below~880MHz the distance of SP antennas is~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™ system. Figs.8,9 compare measured directivities (solid red lines) and simulations (dashed blue lines). As the absolute directivity is difficult to measure, the measured radiation patterns 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 2.8dB for the FLG inductive loop antenna and 1.7dB 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, 2.15dBi (decibels relative to isotropic radiator)[50]. This can be attributed to the asymmetry of the transponders combined with the FLG R_S .

CONCLUSIONS

UHF RFID transponders with screen-printed and sprayed FLG antennas were designed, fabricated and tested. Read ranges~6.7 and 11.1m were measured for antennas with FLG inductive loop and 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.

ACKNOWLEDGEMENTS

We acknowledge funding from EU Graphene Flagship, the French RENATECH network, ERC Grants Hetero2D and MINERGRACE, EPSRC Grants EP/K01711X/1, EP/K017144/1, EP/N010345/1, and EP/L016087/1.

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