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Carrier mobility up to 10^6 cm^{2}V^{-1}s^{-1} measured in a single-crystal diamond by the time-of-flight electron-beam-induced current technique

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Diamond is a very promising material for various applications, and understanding its basic properties is key for the development of future devices. In particular, the low-field mobility of holes has never been measured below ∼80 K in ultra-pure diamond. In order to measure this mobility, we developed an innovative Time of Flight electron Beam Induced Current (ToF-eBIC) technique. A 1 ns pulsed electron beam was made to impact the diamond semiconductor, inducing charge carrier creation and motion through the 546 µm thick bulk diamond, under the influence of an applied electric field. The resulting signal was analysed using the transient time technique. Thus, the velocity of electrons and holes was assessed as a function of the temperature from 13 to 300 K and as a function of the electric field with values ranging from 1.5 to 9200 V/cm. A low-field mobility value of $1.03 \pm 0.05 \times 10^6$ cm^2 V^{-1} s^{-1} was measured for holes at 13 K, demonstrating that diamond is a suitable material to transport charge carriers in a ballistic regime at a scale of ten micrometers.

I. INTRODUCTION

Diamond single crystal is a fascinating material because of ultra wide band gap, large breakdown field, radiation hardness, and excellent charge transport properties. These properties mean diamond can be used in varying fields, including particle detection [1], power electronics [2], valleytronics [3, 4], and so forth. Among diamond's aforementioned properties, charge carrier mobility is perhaps the most relevant property for many applications. Electron and hole mobilities in ultra-pure diamond have been thoroughly investigated over the last four decades by different Time of Flight (ToF) techniques [3, 5–15], also called Transient Current Techniques (TCT), and by Time Resolved and Continuous-Wave Cyclotron Resonance (TRCR [16, 17] and CWCR [18]).

Very high electron mobility was reported by both TCT [15] and cyclotron resonance techniques [16–18]. Recent CWCR experiments enabled researchers to assess electron mobility values up to 100×10^6 cm^2 V^{-1} s^{-1} at 3 K [18], which is 16 times higher than previously reported TRCR values. However, the low-field mobility of holes has never been measured in ultra-pure material by the TCT, at temperatures below ∼80 K. Although some previous studies have reported a saturation of the hole low-field mobility of around 10^4 cm^2 V^{-1} s^{-1} at 2 K because of neutral impurities within the crystal [14], other experiments found that it is necessary to reach extremely low electric fields to perform these measurements below ∼80 K in ultra-pure diamond [13]. As a result, the highest hole low-field mobility reported using the TCT is between 1 and 2×10^4 cm^2 V^{-1} s^{-1} [6, 11] at about 80 K. Using the TRCR technique, hole drift low-field mobility values of 2.3×10^6 cm^2 V^{-1} s^{-1} for light holes and 2.4×10^5 cm^2 V^{-1} s^{-1} for heavy holes at 10 K were measured in 2014 [16]. These mobility values allow us to consider mean free paths of more than 10 micrometers, which are particularly important for the realization of large-scale quantum devices requiring coherent and thus ballistic transport between distant qubits, i.e. color centers in diamond [19, 20]. However, these mobility values were determined by using the momentum scattering time assessed from the cyclotron resonance line width in a region delineated by the cyclotron radius of the measured carriers. In this context, a large-scale distance measurement, as achieved through drift velocities by ToF techniques where the carriers pass through the entire thickness of a several hundred micrometers thick sample, is necessary to demonstrate the possibility of achieving large-scale ballistic transport.

In the various ToF measurements, different particles were used in order to generate electron-hole pairs just below an electrode, such as α particles [1, 8, 10, 14, 21, 22], pulsed X-rays [23], UV light [3, 11–13] and accelerated pulsed elec-

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trons [6, 24]. Then the current induced by the drift of one type of charge carrier under an applied electric field was measured as a function of time. The other type of carriers was collected almost immediately. This phenomenon can be described by the Shockley-Ramo theorem [25, 26].

In this work, we propose a low temperature ToF technique using a pulsed electron beam to generate carriers, which induce a current (eBIC) under an applied electric field. A similar experiment was developed by Quaranta *et al.* [24] and used to study hole transport properties at various temperatures [6]. Our ToF-eBIC technique allowed us to monitor multiple properties independently and simultaneously: 1) our experiment determined the in-depth distribution of electron-hole pairs close to the contact by varying the energy of the electron beam from 500 eV to 30 keV (penetration depth up to $\sim$5 µm); 2) our experiment monitored the electron-hole pairs generated at different electron beam currents ranging from 4 pA to 100 nA during the pulse; 3) our method examined the lateral distribution of charge carriers within a pulse and during all the acquisition time through the focus and the deterministic positioning of the beam, respectively. Thanks to this added freedom and our use of a careful method to avoid the polarization effect in high quality and ultra-pure diamond, we successfully measured the low temperature velocity for electric fields as low as 1.5 V/cm for holes and 18 V/cm for electrons.

The first section of this report describes the experimental setup and the originality of the method. The second section analyzes the ToF-eBIC traces of electrons and holes and assesses the drift velocity versus electric field and temperature. Then, the low-field mobility values of both carriers is reported versus temperature, demonstrating that values up to $10^6 \mathrm{cm}^2\,\mathrm{V}^{-1}\,\mathrm{s}^{-1}$ were achieved in the case of holes.

II. EXPERIMENTAL DETAILS AND METHOD

A 546$\pm$4 µm thick single crystal diamond, grown through chemical vapor deposition (scCVD) and with an electronic grade quality ([N] $< 9 \times 10^{14}\,\mathrm{cm}^{-3}$, typically 0.2 to $2 \times 10^{14}\,\mathrm{cm}^{-3}$, and [B] $< 2 \times 10^{14}\,\mathrm{cm}^{-3}$) from Element 6 has been investigated. The bottom and upper sides of the diamond were metalized using laser lithography and vapor deposition systems. The metallization was composed of a 10/100 nm bilayer of Titanium/Aluminium.

The ToF-eBIC setup proposed here (Fig. 1a,b) is based on the architecture of a scanning electron microscope (SEM). The SEM was a FEI Inspect F50 fitted with an in-house beam blanker driven by a 70 ps rise and fall time pulse generator (Pulse Rider PG-1072 from Active Technologies). The pulses of electrons are generated with a full width at half maximum of 1 ns at a frequency of 333 Hz. The sample was biased using voltage values ranging from -500 to 500 V. The readout system was composed of one CIVIDEC C2HV current preamplifier with a 2 GHz bandwidth connected to a 40 GSa Digital Sampling Oscilloscope (DSO) Lecroy HDO 9404. It was possible to cool the sample to 13 K with a helium liquid cryogenic stage (Gatan CF302) equipped with a thermal shield. Silver paste was used to attach the bottom side of the diamond to the cryogenic stage (side electrically grounded) and to connect a thermalized cable on its upper side. To obtain an accurate and real-time reading of the temperature of the sample, a calibrated CERNOX sensor CX-1070-S-HT-4L was mounted close to the diamond.

To obtain a high signal-to-noise ratio, the electron-beam energy was set at 30 keV to generate a charge of about 11 fC per pulse in the diamond device. During the 1 ns electron-beam pulse, around 30 primary electrons collided with the diamond surface covered with a 110 nm thick Ti/Al contact layer. A 33 ps average time separated each primary electron impact, which generated an average of 2200 electron-hole pairs during the electron-matter interaction within a time frame less than 1 ps, in a pear shaped volume 5 µm deep and 4 µm wide (see Fig 1c).

However, to avoid local effects especially at a low applied electric field (i.e. when electrons and holes generated during consecutive impacts mixed together because of their low drift velocities), the primary electron-beam was unfocused to form a spot of 60 µm in diameter (see Fig 1c). Under these conditions, each primary electron of a single pulse interacted in its own pristine volume, free of charges from previous impacts. This allowed us to minimize polarization effects, i.e. internal electric field induced by the trapping of charge carriers, and carrier-carrier interactions, i.e. electron-hole recombination rate and local electric field screening.

Moreover, after a high energy irradiation, a mixture of excitons and free-carriers is generated. Due to the large binding energy of excitons in diamond, the fraction of excitons of the mixture at equilibrium is almost equal to one at low temperatures [27]. Conversely to free charge carriers, the excitons are neutral particles and thus, are not impacted by the electric field which could lead us to conclude that the ToF-eBIC technique cannot be used at low temperatures. Nevertheless, by applying an electric field, the electrons and the holes drift in opposite directions and can thus be separated prior to the formation of excitons. This effect is limited when the excitation density is high (around 10 nm focused electron beam in Fig 1c) due to the difficulty of separating the charge carriers in a large and dense cloud of electrons and holes. Again conversely, for low carrier density (unfocused electron beam in Fig 1c), this phenomenon is sufficient to decrease the quantity of excitons generated significantly.

III. TEMPERATURE DEPENDENCE OF THE DRIFT VELOCITY AND CARRIER MOBILITY

The ToF-eBIC traces shown in Fig. 2 were obtained by averaging single waveforms taken at different positions as the beam was scanned over an area allowing the spatial separation of pulses (typically 200 µm apart). Once the area was scanned, the acquisition was paused and then to reset the state of the diamond, a cycling procedure was performed by inverting the bias polarity for 1 s while keeping the electron beam excitation constant. Finally, the acquisition process was resumed. Acquisition/cycling procedures could be repeated 100 times for the lower electric fields to obtain waveforms in which the

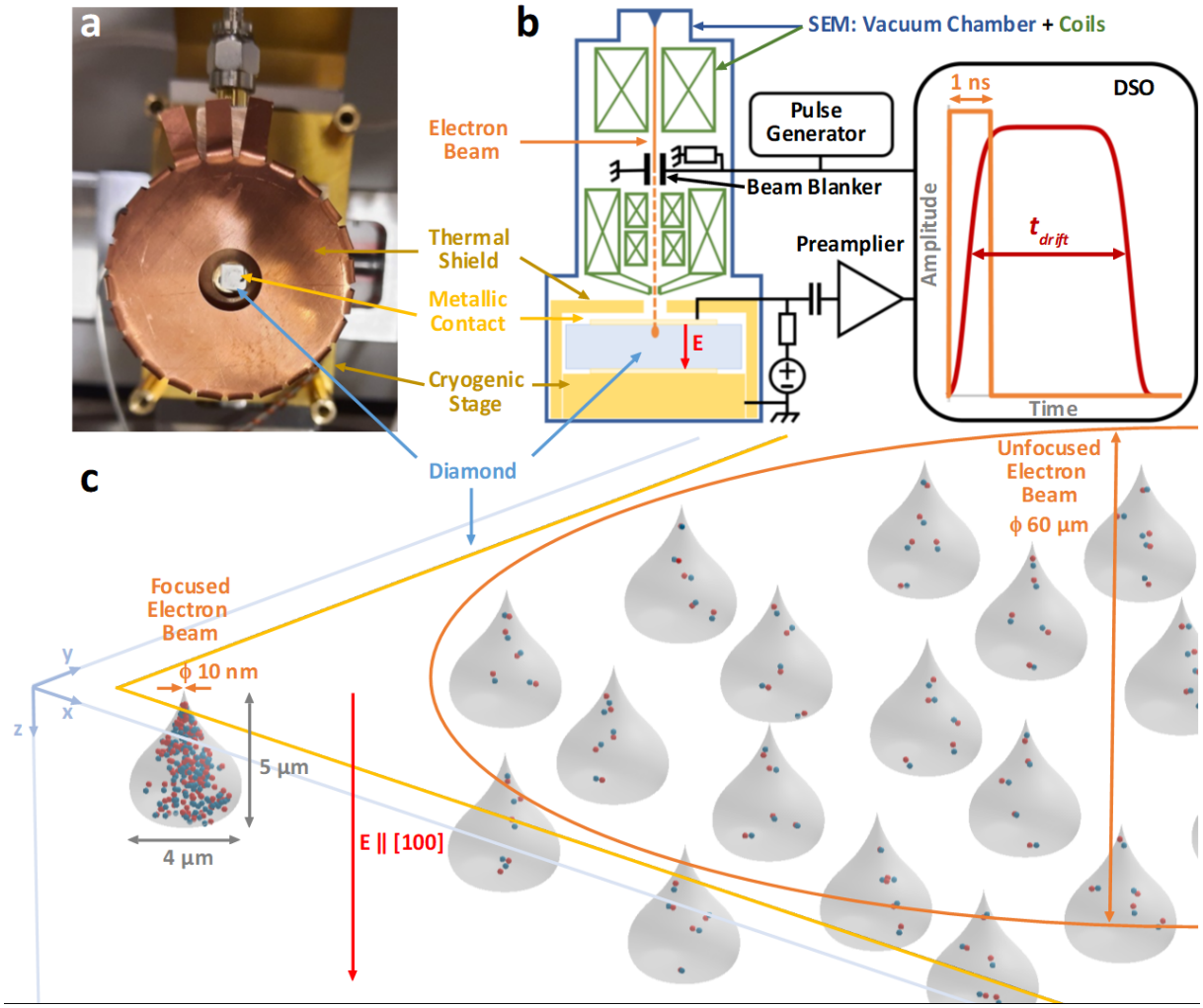

FIG. 1. Time of flight electron beam induced current setup. a) Top view picture of the diamond sample on the cryogenic stage surrounded by the copper thermal shield. b) Schematic of the e-beam set up, the signal conditioning, and the digital storage oscilloscope (DSO) treatment. The orange curve of the chronogram corresponds to the electron beam current pulse controlled by the beam blanker. The red curve corresponds to the current passing through the diamond, induced by the applied electric field E, and processed by the preamplifier. c) Schematic of the electron-hole pair (respectively the blue and red dots) populations created within a focused electron beam and an unfocused electron beam. The orange line delineates the lateral size of the unfocused beam and the yellow line the outer limit of the metallic contact through which the beam excites the electron hole pairs in diamond. The gray pear-shaped volumes represent the random lateral distribution of the regions, where the electron-hole pairs may be created by each primary electron impacting the sample surface.

rising and falling edges of the signal could be clearly identified.

These traces for electrons and holes were obtained at different temperatures and different applied electric fields. Multiple conclusions can be drawn from these results. First, the transit times are systematically shorter for holes than for electrons, across all the electric fields and temperatures. Secondly, the transit time of both carriers increases with the temperature for a given electric field (except for the electrons transits measured at 120 and 150 K), demonstrating that the drift velocity decreases with temperature (see also Fig. 3a-d) with a maximum value measured at 13 K for a given electric field. For temperatures above 150 K, the signal shapes of electrons and holes are very similar. However, for temperatures below 150 K and for low electric fields (E < 500 V/cm), the electron signal differs fundamentally from the signal of holes with traces accounting for two steps. Indeed, for these traces, the transits of the electrons in the longitudinal (cool electrons) and transverse (hot electrons) valleys can be studied separately. The first contribution of the signal was attributed to the hot electrons which have a lower effective mass than the cool electrons [28]. Valley-polarized electrons were experimentally observed in diamond samples and theoretically described by Isberg *et al.* in 2013 [3].

To estimate the drift velocity, the method described by Isberg *et al.* [30–32] (see also the Supplemental Material [33] for the evaluation of the drift velocity errors) was used to reproduce $I(t)$, the amplitude of the trace versus time of Fig.

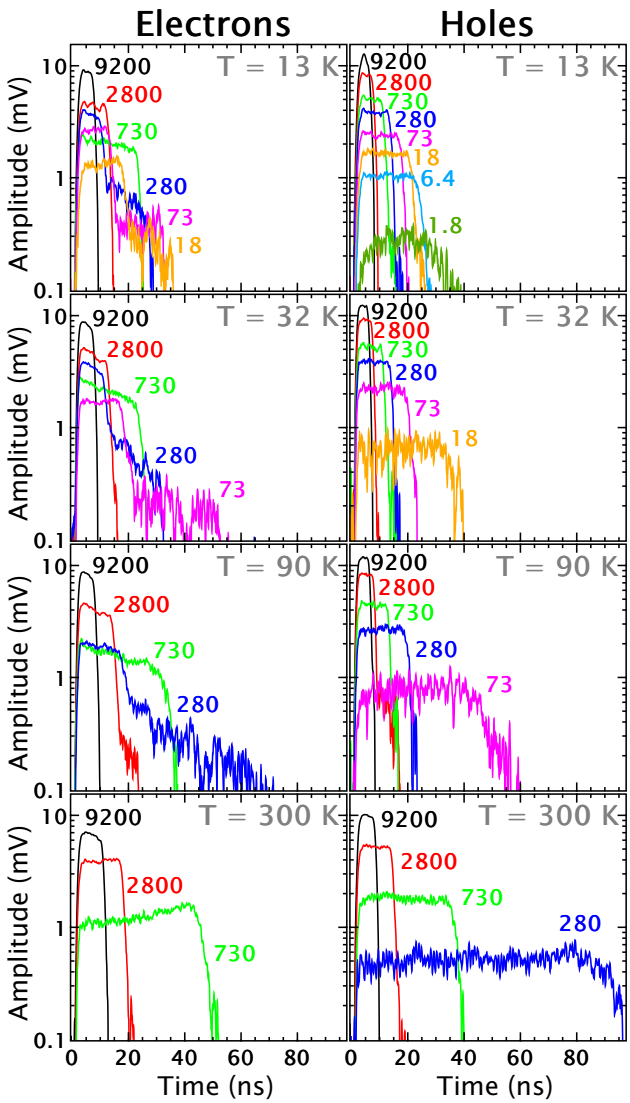

FIG. 2. Time of flight traces of electron and hole. Time of flight traces measured at different temperatures (T=13, 32, 90 and 300 K) and electric fields. The electric field in V/cm is specified in a label next to each curve.

2, and extract its full width at half maximum as well as the experimental uncertainties [34]. In Fig. 3a-d, the drift velocities of holes and electrons versus the electric field are shown for temperatures between 13 and 300 K. For most cases, it is remarkable that an ohmic behavior was observed at the lowest electric field and thus that the low-field mobility μ_0 values could be extracted by a linear fit (solid lines).

The hole and electron low-field mobilities were estimated at $2190 \pm 30\,\mathrm{cm^2\,V^{-1}\,s^{-1}}$ and $1660 \pm 10\,\mathrm{cm^2\,V^{-1}\,s^{-1}}$ (respectively) at room temperature. These values are consistent with the low-field mobilities, measured using the TCT, reported in the literature[1, 10, 11, 21, 23]. Fig 3c shows that at 120 K, the electron drift velocity decreases between 360 and 550 $\mathrm{V\,cm^{-1}}$. This phenomenon is called negative differential mobility and, it was observed for the first time in a diamond sample by Isberg *et al.* in 2012 [12]. Our measurements accurately reproduce their results obtained at 120 K which demonstrates the reliability of our method. For temperatures greater than 220 K (Fig 3c,d), a fit within the whole electric field range was also performed using equation (1) (dashed line) [35].

$$v_{drift}(E) = \frac{\mu_0 E}{1 + \frac{\mu_0 E}{v_{sat}}}, \tag{1}$$

where v_{drift} is the drift velocity, E the applied electric field, μ_0 the low-field mobility, and v_{sat} the saturation drift mobility. This fit function accurately describes the electric field dependence of the drift velocity at high temperatures (higher than ~200 K).

The established empirical model described by Caughley and Thomas [35] cannot be used for holes at the lowest temperatures as reported by Majdi *et al.* [13]. Indeed, at low temperatures, the accelerating electric field leads to the carrier temperature becoming substantially larger than the lattice temperature, inducing a deviation from the ohmic behavior. As expected, the threshold of the electric field delineating the ohmic behavior to the hot carrier behavior increases with temperature. Very low-fields are needed to assess the mobility values at low temperature (see Supplemental Material [36] at [URL will be inserted by publisher] for the fitting method and uncertainties). For example, electric fields lower than 10 V/cm are needed for temperatures lower than 30 K. Our optimized experimental method enabled us to maximize the signal to noise ratio and thus measure the ToF-eBIC signals at extremely low-fields reaching the ohmic region.

The temperature dependence of the low-field mobility of electrons and holes is shown in Fig. 3e. It is important to note the extremely high mobility value of holes measured in this work $\mu_0 = 1.03 \pm 0.05 \times 10^6\,\mathrm{cm^2\,V^{-1}\,s^{-1}}$ at 13 K. This mobility can be compared with the hole mobility value of $6.4 \times 10^5\,\mathrm{cm^2\,V^{-1}\,s^{-1}}$ averaged from the light and heavy hole mobility values ($2.3 \times 10^6\,\mathrm{cm^2\,V^{-1}\,s^{-1}}$ and $2.4 \times 10^5\,\mathrm{cm^2\,V^{-1}\,s^{-1}}$, respectively) at 10 K from Akimoto *et al.* [16]. Considering the heavy and light hole bands separately (see Supplemental Material [37] at [URL will be inserted by publisher] for evaluation method of the light and heavy holes mobility values), the light hole mobility $\mu_{lh} \cong 3.62 \times 10^6\,\mathrm{cm^2\,V^{-1}\,s^{-1}}$ and heavy hole mobility $\mu_{hh} \cong 3.88 \times 10^5\,\mathrm{cm^2\,V^{-1}\,s^{-1}}$ values have been evaluated from this work.

The average mobility value measured in the present work is the highest value reported for holes in a bulk semiconductor, approaching the hole mobility of 2D GaAs systems $5.8 \times 10^6\,\mathrm{cm^2\,V^{-1}\,s^{-1}}$ measured at 0.3 K [38] or graphene $1.4 \times 10^6\,\mathrm{cm^2\,V^{-1}\,s^{-1}}$ measured at 1.8 K [39].

The hole mobility values reported here are also higher than those of electrons in diamond measured by UV-ToF[17] as shown Fig. 3e. However, Konishi *et al.* recently reported electron mobility values of up to $100 \times 10^6\,\mathrm{cm^2\,V^{-1}\,s^{-1}}$ measured by CWCR, a local probe technique, at 3 K [18]. Mobility values measured by cyclotron resonance have been explained to be higher than macroscopic measurements due to crystalline inhomogeneity [17].

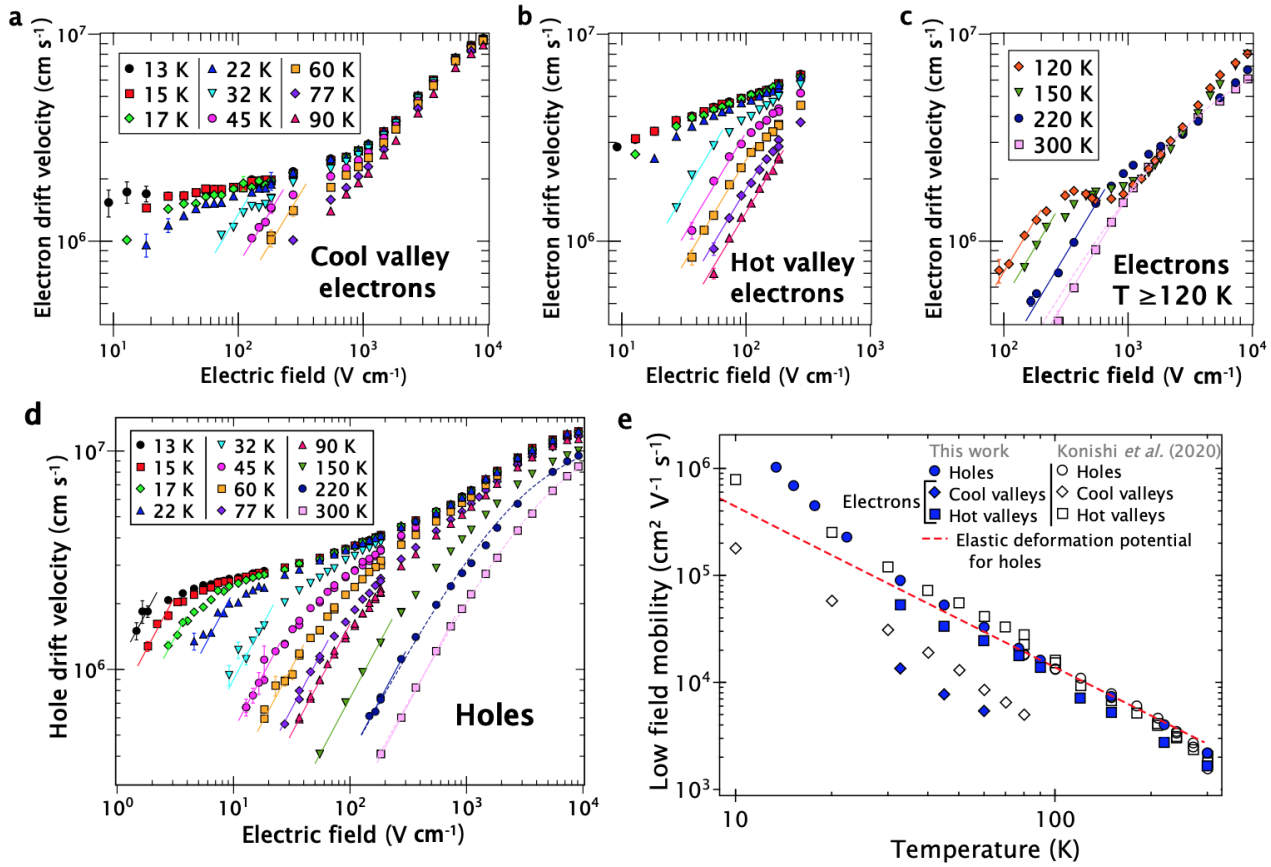

FIG. 3. Electron and hole drift velocities and low-field mobilities. **a,b**, Cool valley (**a**) and hot valley (**b**) electron drift velocity versus electric field at various temperatures. The temperatures in **b** are indicated in the legend in **a**. **c**, Electron drift velocity versus electric field measured above 120 K. **d**, Hole drift velocity versus electric field at various temperatures. The solid lines show the linear fit at low electric fields used to extract the low-field mobility. A fit on the entire electric field range was also performed using eq. (1) at 220 and 300 K (dashed lines). **e**, Hole and electron low-field mobility versus temperature. The blue markers correspond to the values extracted during this work and the open markers to the value of Konishi *et al.* [17]. The red dashed line represents the theoretical low-field mobility expected for holes based only on the elastic deformation potential extracted from Pernot *et al.*[29].

For temperatures higher than T=30 K, a $T^{-3/2}$ mobility dependence was observed. This dependence was expected because of the acoustic phonon scattering of holes, which is the main limitation for ultra-pure material in this temperature range. Calculations were performed to evaluate the hole low-field mobility at low temperatures [29], and the theoretical results were compared to the experimental data. It is important to note that the theoretical hole low-field mobility thus calculated does not take the inelastic acoustic phonon scattering of holes into account. As a result, for T $<$ 30 K, a discrepancy between the elastic model and the experimental data could be observed. Indeed, the inelastic scattering phenomenon leads to higher hole low-field mobility. Therefore, if the crystal can be considered sufficiently pure to the extent that the elastic scattering caused by impurities can be neglected, hole low-field mobility values comparable to the ones measured for electrons ($100 \times 10^6\,\mathrm{cm^2\,V^{-1}\,s^{-1}}$ at 3 K [18]) are expected at a temperature of few Kelvins. In any case, the light and heavy hole mobility values extracted here correspond to a relaxation time of $\tau_{lh} \cong 537$ ps and $\tau_{hh} \cong 146$ ps and mean free paths of $l_{lh} \cong 25$ µm and $l_{hh} \cong 4.3$ µm (see Supplemental Material [37] at [URL will be inserted by publisher] for evaluation method of the light and heavy holes relaxation times and mean free path). The mean free path of light holes is around 20 times shorter than the 546 µm gap between electrodes meaning that the transport regime is quasi-ballistic for such carriers.

The hole mean free path larger than 10 µm at a macroscopic scale above liquid helium temperature (T = 4 K) provides new opportunities to transport quantum information and preserve its coherence between two qubits in diamond. Recently, Lozovoi *et al.* [19] proposed a method based on confocal fluorescence microscopy and magnetic resonance to induce and probe charge transport between individual nitrogen-vacancy centers in diamond at room temperature. The same group argued that the use of an external electric field and the lowering of temperature should drastically improve the hole transport fidelity due to a ballistic regime on a scale comparable with the inter-qubit distance [20]. Our work demonstrates the possibility of reaching ballistic regime at this scale and provides accurate data concerning the achievable distance/temperature

trade-off needed to increase the transport fidelity.

IV. SUMMARY

In summary, thanks to the implementation in a SEM of a time-of-flight technique based on electron pulses to generate carriers in a bulk diamond, we were able to measure the electron and hole drift velocities from the transient currents down to 13 K for a large range of electric field (1.5 to 9000 V/cm). The low-field mobility of the charge carriers was extracted and compared with the literature. At 13 K, the low-field mobility of holes was found to reach a remarkable value of $1.03 \pm 0.05 \times 10^6\,\mathrm{cm}^2\,\mathrm{V}^{-1}\,\mathrm{s}^{-1}$. This value corresponds to the highest averaged hole low-field mobility measured in a bulk semiconductor.

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