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Low-temperature mobility-lifetime product in synthetic diamond

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Mobility-lifetime ($\mu\tau$) product is an important parameter that determines the performance of electronic and photonic devices. To overcome the previously reported difficulties in measuring the $\mu\tau$ product at cryogenic temperatures, we implement a time-resolved cyclotron resonance method to determine the carrier lifetime τ . After clarifying the difference between the AC and DC mobilities measured by the cyclotron resonance and time-of-flight methods, respectively, we demonstrate an inverse temperature dependence of the $\mu\tau$ product. The highest recorded $\mu\tau$ product of 0.2 cm²/V, which is approximately 100 times the room-temperature value, was obtained at 2 K for chemical-vapor-deposition diamond of the highest currently available purity.

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Diamond is an attractive material for high-power electronics and photonics owing to its extraordinarily high thermal conductivity, breakdown voltage, and mobility^{1–3}. In addition, the high radiation hardness and long carrier lifetime^{4,5} originating from the indirect bandgap make diamond an ideal material for high-sensitivity radiation detectors. The performance of a diamond radiation detector^{6–8} depends on both the carrier mobility and lifetime. Therefore, the charge collection efficiency is evaluated using the mobility-lifetime ($\mu\tau$) product, i.e., the product of the carrier mobility μ and lifetime τ . Knowledge of the $\mu\tau$ product is also essential for evaluating the photoelectric gain for the spin readout of single nitrogen vacancy centers, which is useful for quantum information processing^{9,10}.

The recent implementation of a diamond detector at cryogenic temperature, as a beam-loss monitor in the Large Hadron Collider at CERN¹¹, has attracted a considerable attention for the $\mu\tau$ product at low temperature. The most common experimental approach to the $\mu\tau$ product is to measure the charge collection and analyze the voltage dependence using the Hecht model¹². Previous studies have evaluated the $\mu\tau$ products in polycrystalline¹³ and monocrystalline diamonds at 300 K¹⁴. The value ($\mu\tau = 3.3 \times 10^{-3} \text{ cm}^2/\text{V}$) in monocrystalline diamonds was 2–3 orders of magnitude greater than that in polycrystalline diamonds affected by carrier trapping at grain boundaries. This result indicates the high potential of monocrystalline diamonds among other candidate materials, such as high-purity germanium, microcrystalline silicon¹⁵, CdZnTe¹⁶, metal halide perovskites¹⁷, and h-BN¹⁸. However, the charge collection method returns the product of μ and τ , and these two parameters are not separable. Further, the knee bias voltage characterizing the $\mu\tau$ value becomes extremely small at temperatures below 80 K, which makes it difficult to determine the $\mu\tau$ product at cryogenic temperatures.

In this study, we propose to evaluate the $\mu\tau$ product by measuring μ and τ separately using other techniques, that is, the time-resolved cyclotron resonance (TRCR) and time-of-flight (ToF) methods. In contrast to standard electrical measurements such as Hall effect measurement, the TRCR method generally provides two relevant transport parameters, i.e., the carrier mobility and lifetime, from a single measurement¹⁹. However, the mobility measured by the TRCR method was found to be considerably higher than that measured by the ToF method. By comparing the TRCR and ToF mobilities of the same samples, we clarify the difference between the AC and DC transport properties. Consequently, our analysis reveals the inverse temperature dependence of the $\mu\tau$ product down to cryogenic

temperatures and the effect of a residual strain as small as 10^{-6} . In this paper, after describing the experimental setup, we discuss the mobility, lifetime, and mobility-lifetime product.

The three single-crystal diamonds (samples 1 – 3) of the highest purity currently achievable were grown by the chemical vapor deposition (CVD) method at Element Six. The impurity concentration in sample 3 is 0.05 ± 0.03 ppb for nitrogen and below 0.2 ppb for boron. The estimated capture times of electrons at neutral nitrogen and boron impurities are $\tau_N = 1$ ms and $\tau_B > 1.5$ μ s at 10 K²⁰, which are longer than the measured decay times presented below. Each sample has an inhomogeneous strain distribution on the order of 10^{-5} , which was assessed and reported in Ref.²¹. The strain map of sample 2 is shown in Fig. 1. The spatial inhomogeneity of the strain results in a position-dependent excitation lifetime²¹. The TRCR spectra were obtained by measuring the transmission of X-band microwave at $\omega/2\pi = 9.64$ GHz under a magnetic field with uniform photoexcitation of carriers over the sample area by laser light (pulse width: 5 ns, wavelength: 225 nm). The resonance magnetic field B_0 , half linewidth Γ , and amplitude A were obtained by Lorentzian fits to the spectra at each delay time³. The signal intensity I was obtained by multiplying the width and amplitude of the resonance peak (i.e., $I = 2A\Gamma$), and the linewidth in the large time delay limit was used to avoid the carrier-carrier interaction effect in extracting the mobility. As the carrier lifetime is longer in less strained regions than in more strained regions, the TRCR mobility values are considered to provide information from the regions with the highest crystallinity in the samples. To obtain the ToF mobility, carriers were photoexcited by 800 ps pulses at a wavelength of 213 nm and measured using Ti/Al mesh contacts²². The drift mobility was derived from the velocity data obtained under an electric field applied along the [100] axis of the crystal. The charge collection measurements were conducted under 224 nm excitation with 100 μ s pulses and using Ti/Al and Au electrodes.

An example of the CR spectra of sample 1 at 4 K is shown in the inset of Fig. 2. Three sharp peaks (marked e1, e2, and lh) and one broad peak (hh) represent resonances resulting from different species of carriers with different effective masses²³. Owing to the low carrier temperature realized in this study, the spectral overlap of the resonances observed in previous studies^{3,23} was completely resolved. Using a Lorentzian fit (solid line), the scattering time was extracted as $t_p = B_0/(\omega\Gamma)$ and the mobility was determined as $\mu = qt_p/m$, with a carrier effective mass $m = qB_0/\omega$ and the elementary charge q . The circles in Fig. 2 show

the mobility obtained from the e1 resonance at $B_0=122$ mT ($m_1 = 0.35m_0$) in samples 1–3. We recorded the highest mobility ever reported for diamond (approaching 10^7 cm²/Vs) by lowering the carrier temperature and density in these crystals with remarkably low impurity concentrations. However, it was impossible to extract the mobility at temperatures above 40 K because of the broadening and overlap of the CR spectral peaks.

The triangles and squares in Fig. 2 show the drift mobility measured by the ToF method. The data for cool electrons in valleys on the [100] axis with longitudinal mass ($m_l = 1.56m_0$ ²³), hot electrons in valleys on the [010] and [001] axes with transverse effective mass ($m_t = 0.28m_0$ ²³), and holes are plotted using different symbols. The measurement was difficult below 10 K owing to carrier heating by the applied electric field. Thus, both TRCR and ToF mobility measurements were possible from 10 to 40 K. In this temperature range, the ToF mobility values are approximately one-fourth those determined by the TRCR method considering the effective mass difference. A similar difference has been observed between the cyclotron (AC) and transport (DC) mobility in other carrier systems²⁴, and can be explained as follows. The cyclotron orbits of carriers under microwave at 9.64 GHz are approximately 100 nm. This means that the TRCR measurement probes carriers' AC motion in a minuscule region compared to the whole sample volume. In addition, as we extract the scattering time at long delay times, the dominant contribution in the TRCR mobility arises from the carriers surviving in high-quality areas of the sample (Fig. 1), where residual strain is remote and scattering is suppressed. In contrast, the ToF mobility measures the averaged DC behavior of carriers propagating through the sample with crystalline inhomogeneity. As a higher scattering rate is experienced in lower quality areas of the sample, the ToF mobility is measured naturally lower than the TRCR mobility.

In diamond, the mobility is dominated by acoustic phonon scattering¹, where the scattering rate varies with temperature T as $T^{3/2}$ in the elastic approximation. We note that the TRCR mobility values below 10 K are well reproduced by adding a temperature-independent scattering term, b , to the acoustic phonon scattering rate γ_{ac} :

$$\mu_{CR} = \frac{q}{m_1}(\gamma_{ac} + b)^{-1}. \quad (1)$$

The solid line in Fig. 2 represents Eq. (1) with $q/(m_1b) = (6.1 \pm 0.4) \times 10^6$ cm²/Vs. The dash-dotted line is an empirical fit to the ToF mobility for electrons with the conductivity

$$\text{mass } m_c = 3/(2/m_t + 1/m_l) = 0.385m_0,$$

$$\mu_{\text{ToF}} = \frac{q}{m_c}(3.7\gamma_{ac} + b)^{-1}. \quad (2)$$

According to the literature²⁵, the rate of acoustic-phonon scattering is dependent on the microwave frequency, whereas the impurity scattering rate is independent of the frequency. By analogy, we assumed that b is independent of frequency and is common to the ToF and TRCR values.

Next, we present the temperature dependence of the carrier lifetime. The temporal variation of the electron (e1) TRCR signal in sample 1 at 3–300 K is shown in Fig. 3(a). For temperatures above 80 K, we simply counted the signal intensity centered at B_0 because the CR spectra broadened and separation of the carrier species was difficult. The signal decayed double exponentially. The faster decay is attributed to many-body effects at a high electron density immediately after photoexcitation³. Therefore, we extracted the time constant τ of the slower decay by fitting the data with the sum of exponential rise and decay, $I(t) = I_0[-\exp(-t/\tau_0) + \exp(-t/\tau)]$. The fitting functions are indicated by dashed lines. The lifetime tends to be longer at higher temperatures. Figure 3(b) compares the decays measured in each sample at 10 and 300 K. The electron lifetime was extracted as $\tau_1 = 500$ ns, $\tau_2 = 350$ ns, and $\tau_3 = 170$ ns, respectively, for samples 1, 2, and 3 at 10 K. The closed circles in Fig. 3(c) show the electron lifetime as a function of temperature. The relation $\tau_1 > \tau_2 > \tau_3$ was found below 40 K. Surprisingly, the samples showed different lifetimes, although they showed similar mobility values. In the following, we discuss the reasons for the variation of the lifetime depending on the sample and temperature.

As the lifetimes are not limited by capture at impurity sites at extremely low concentrations²⁰, we consider the effect of increasing mobility with decreasing temperature. The mobility is related to the diffusion coefficient D by the Einstein relation, $D = \mu k_B T / q$, where k_B is the Boltzmann constant. A higher mobility indicates a higher diffusivity, which may result in a decreased lifetime as the carriers diffuse to the recombination centers. The carrier decay time associated with surface recombination is given by the surface lifetime²⁶:

$$\tau_s = \frac{d}{2S} + \frac{1}{D} \frac{d^2}{\pi^2} = \frac{d}{2S} + \frac{q}{\mu k_B T} \frac{d^2}{\pi^2}, \quad (3)$$

where S is the surface recombination velocity. As in the case of exciton decay reported in Ref.²¹, we replace S with the modified surface recombination velocity S' and the sample

thickness d with the effective distance between dislocations, $2w$, at which carriers nonradiatively recombine. Here, we use μ_{ToF} instead of μ_{CR} for μ in Eq. (3), considering that the DC mobility, rather than the AC mobility, is relevant for the migration of carriers to a dislocation²⁴. The best fits, as shown by the solid lines in Fig. 3(c), were obtained for $S' = 0.13 \times 10^6$ cm/s²¹, $w_1 = 0.24$, $w_2 = 0.19$, and $w_3 = 0.15$ mm. The effective distances were shorter than the sample thickness of $d \simeq 0.5$ mm, and they can be converted to strain magnitudes of $\epsilon_1 = 2.3 \times 10^{-6}$, $\epsilon_2 = 3.5 \times 10^{-6}$, and $\epsilon_3 = 5.5 \times 10^{-6}$, using the relation recently established between the effective distance for excitons and strain²¹. According to the actual strain map in Fig. 1, it is likely that these values correspond to the strains in the regions with the best crystallinity.

In Fig. 3(c), the data below 40 K and the calculation according to Eq. (3) are in good agreement. This indicates that a small strain does not affect the mobility, but does limit the lifetime. A similar situation has been discussed for the minority carriers in GaAs against dislocation density²⁷. The discrepancy between the data and model above 80 K is attributed to contributions from excitons. At these temperatures, the excitons start to thermally ionize to free carriers according to the mass action law²⁸. This leads to the formation of a gas mixture of electrons, holes, and excitons whose treatment is challenging. To provide insight, the dotted lines represent the expected lifetime of excitons in each sample, which are determined mainly by their radiative recombination²¹.

Next, we discuss the $\mu\tau$ product. The collected charge Q was measured for varying bias voltages V , and analyzed using the Hecht expression¹²:

$$Q = N_0 q \frac{\mu\tau V}{d^2} \left[1 - \exp\left(-\frac{d^2}{\mu\tau V}\right) \right]. \quad (4)$$

We convolved Eq. (4) with the Beer–Lambert law to accommodate for a light penetration depth of ~ 200 μ m and fitted it to the data to obtain the $\mu\tau$ product. Here, $N_0 q$ is the total charge generated and $d = 0.5$ mm is the sample thickness. The fit for 120 K is shown as an example in the inset of Fig. 4. The $\mu\tau$ products thus obtained ranged from 10^{-2} to 10^{-4} cm²/V at 80 – 300 K.

In radiation detectors, the charges are measured under an external voltage applied across the intrinsic diamond layer, with electrodes covering a broad region of the front and back sample surfaces. Thus, the measured $\mu\tau$ product is an average over the sample area, where the carrier lifetime is expected to depend on the position owing to inhomogeneous strain. To

compare this with the TRCR results obtained from the regions of highest crystallinity, we corrected the TRCR lifetime by multiplying them by the ratio of the smallest and average strain magnitudes, i.e., $\epsilon/\tilde{\epsilon}$. This correction is based on the fact²¹ that w scales as $\epsilon^{-1/2}$, and τ scales as $1/\epsilon$. Here we use the strain magnitude averaged over the circular area of 3 mm in diameter considering the electrode size, $\tilde{\epsilon}_1 = 1.4 \times 10^{-5}$, $\tilde{\epsilon}_2 = 2.0 \times 10^{-5}$, and $\tilde{\epsilon}_3 = 5.6 \times 10^{-5}$. The closed circles in Fig. 4 are products of the electron mobility μ_{ToF} and lifetime τ from the TRCR measurements corrected by factor $\epsilon/\tilde{\epsilon}$. The open circles indicate the products of μ_{ToF} averaged for the electrons and holes and the corrected τ . The dots with error bars show the means of the electron $\mu\tau$ and hole $\mu\tau$ from the Hecht analysis. The agreement between the open circles and dots for each sample demonstrates an important finding; Our estimation is reliable owing to the high accuracy of the strain quantification and relationship established between the strain and effective distance, although it requires some conversions.

The $\mu\tau$ product of electrons below 40 K diverged, reflecting the sample dependence of τ . This result is expected because, in this temperature range, τ is determined by the arrival time owing to diffusion to dislocations. If the first term in Eq. (3) is neglected (which is a reasonable approximation), we obtain

$$\mu\tau_s \left(\frac{\epsilon}{\tilde{\epsilon}} \right) \simeq \frac{q(2w)^2}{k_B T \pi^2} \left(\frac{\epsilon}{\tilde{\epsilon}} \right) \propto \frac{1}{T}. \quad (5)$$

This result clearly indicates that the strain affects the $\mu\tau$ product through the sample-dependent w , ϵ , and $\tilde{\epsilon}$. The expected inverse temperature dependence of the $\mu\tau$ product is shown by the dashed line in Fig. 4. From room temperature to cryogenic temperature, the $\mu\tau$ product varies by two orders of magnitude.

Secroun *et al.* reported that $\mu\tau$ products higher than $10^{-3} \text{ cm}^2/\text{V}$ in CVD diamonds are controlled by a very small number of defects, which cannot be clearly characterized²⁹. Lohstroh *et al.* reported the effect of dislocations for $\mu\tau < 10^{-4} \text{ cm}^2/\text{V}$ ³⁰, and Tarun *et al.* discussed charge collection as a function of the nitrogen-vacancy concentration³¹. We clarified that the $\mu\tau$ product at low temperatures can be modified by a factor owing to the small residual strain on the order of 10^{-6} . Our systematic procedure, relating the AC and DC transport properties and including the spatial inhomogeneity of the samples, confirmed our estimation of the $\mu\tau$ product at 2 K, which had the highest ever value of $0.2 \text{ cm}^2/\text{V}$. The increase in diffusion at low temperature is inherent because of the quenching of carrier-phonon interaction, and the limit of the lifetime due to diffusion is unavoidable. To further

increase the $\mu\tau$ product at cryogenic temperatures, improving the crystallinity to the defect-free level could be effective.

In summary, we investigated the temperature dependence of the carrier lifetime and mobility by combining TRCR, ToF, and charge collection measurements on high-purity diamond, which greatly limited interactions with impurities. The difference between the DC and AC transport properties was explained in terms of crystalline inhomogeneity. The sample dependence was explained by the variation in the lifetime due to the effect of the $<10^{-5}$ residual strain, which has been disregarded in most previous studies. The inverse temperature dependence of the $\mu\tau$ product ($\mu\tau \propto 1/T$) was clearly demonstrated, indicating a new possibility for improving the efficiency of diamond devices.

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DATA AVAILABILITY

The data that support the finding of this study are available from the corresponding author upon reasonable request.

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FIG. 1. Strain map of sample 2, with schematics showing the position-dependent dynamics of electrons. Data adapted from Ref.²¹.

FIG. 2. Temperature dependence of the mobility measured by TRCR and ToF methods. Dashed line represents calculated mobility based on acoustic phonon scattering. Solid and dash-dotted lines include a temperature-independent term [Eqs. (1) and (2)]. Inset: CR spectrum of sample 1 at 4 K. Signal was integrated over delay times of 600 to 1000 ns. Solid line represents a Lorentzian fit.

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FIG. 3. (a) Decay of the e1 signal of sample 1 at various temperatures. (b) e1 signal decay of each sample at 10 K and 300 K. Dashed lines in (a) and (b) represent fitting functions. (c) Comparison of measured decay time (circles) and surface decay times τ_s for electrons (solid lines). Dotted lines indicate exciton lifetimes assuming the same effective distances as for electrons.

FIG. 4. Temperature dependence of the $\mu\tau$ product. Solid circles: electron ToF mobility multiplied by the electron lifetime corrected by strain, open circles: mean of the electron and hole ToF mobilities multiplied by the lifetime corrected by strain, triangles: mean of the electron and hole mobility-lifetime products from charge collection. Dashed line: guide to the eyes showing the $1/T$ dependence. Inset: collected charge (dots) as a function of bias voltage on sample 1 at 120 K and the Hecht fit (solid line).

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