

Quantitative Modeling of the Non-Monotonic Sensitivity of Defect-Engineered Graphene Sensors

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Article

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Abstract

Defect engineering has been explored as a means to improve the performance of graphene-based gas sensors. However, sensor sensitivity often exhibits a non-monotonic relationship with defect density, presenting challenges for device optimization. In this study, we introduce a quantitative physical model aimed at describing and anticipating this behavior. Graphene samples were irradiated with deuteron ions to generate a controlled range of defect densities, which were characterized using Raman spectroscopy. Sensitivity to hydrogen showed a Λ -shaped dependence on defect density, with a peak response observed at an intermediate concentration. This behavior is attributed to the competing effects of signal enhancement and structural degradation induced by defects. The proposed model expresses sensitivity as a function of two experimentally derived parameters an activation coefficient (C_n) and a degradation coefficient (γ_n), providing insight into the underlying sensing mechanism.

Introduction

The development of low-power, room-temperature gas sensors is pivotal for advancing portable environmental monitoring and point-of-care diagnostics. Traditional metal-oxide semiconductor sensors, while offering high sensitivity, rely on elevated operating temperatures, resulting in substantial power consumption and integration challenges.^{1,2} These limitations have accelerated the search for alternative sensing materials, positioning graphene as a highly promising candidate due to its unique physical properties.³⁻⁸ Graphene's atomically thin, two-dimensional lattice endows it with an exceptionally high surface-to-volume ratio, making its electronic characteristics particularly charge carrier density extremely sensitive to ambient conditions.⁹ This sensitivity underpins its chemiresistive sensing behavior, where adsorbed gas molecules modulate electrical resistance by acting as electron donors or acceptors.^{10,11} However, the chemically inert basal plane of pristine graphene, dominated by sp^2 hybridized bonding, weakly interacts with most gas species via van der Waals forces. This weak interaction limits adsorption energy, resulting in modest sensitivity and sluggish response kinetics.¹² To address these intrinsic limitations, defect engineering has emerged as an effective strategy for enhancing surface reactivity. Early studies relied on chemical treatments and plasma exposure to introduce structural imperfections, which improved sensing performance but lacked reproducibility and precision.¹³⁻¹⁷ In contrast, ion beam irradiation a well-established technique in the semiconductor industry offers fine control over defect generation by adjusting ion species, energy, and dosage, enabling systematic modulation of graphene's properties.¹⁸⁻²² Experimental evidence consistently demonstrates that defect-induced activation significantly enhances gas sensitivity.²³⁻²⁵ Yet, the relationship between defect density and sensing performance is non-monotonic: sensitivity improves with increasing defect density up to an optimal point, beyond which it deteriorates.²⁶⁻²⁸ This suggests a trade-off between the creation of reactive sites and the degradation of charge transport pathways. While this phenomenon has been repeatedly observed, the absence of a predictive physical model has hindered rational sensor optimization, leaving researchers reliant on trial-and-error approaches. In this study, we present a physics-based model that attributes the observed non-monotonic trend to two competing physical

effects activation and degradation described by an activation coefficient and a degradation coefficient, respectively. By establishing a direct analytical relationship between sensitivity and defect density, our model aims to transition sensor design from empirical exploration to predictive, physics-based engineering.

EXPERIMENTAL DETAILS

The devices were fabricated on commercial single-layer graphene, grown by chemical vapor deposition (CVD) on a copper foil catalyst and subsequently transferred onto heavily doped silicon wafers with a 300 nm thick thermally grown SiO₂ dielectric layer. The quality and monolayer nature of the transferred graphene were confirmed by Raman spectroscopy. A photolithography step was performed to define the source and drain electrode patterns. Metal contacts consisting of Ti/Au (5 nm/50 nm) were deposited by e-beam evaporation. The prepared devices were irradiated with deuteron (D⁺) ions at the Korea Multipurpose Accelerator Complex (KOMAC), utilizing an electrostatic accelerator. The ion energy was maintained at a constant 120 keV to ensure that the ions fully penetrate the graphene layer and that the primary interaction mechanism is collisional energy transfer rather than implantation. The ion dose was systematically varied from 5×10^{13} to 1×10^{15} ions/cm² in a high-vacuum target chamber (pressure < 5×10^{-6} Torr). To ensure uniform irradiation across the entire device area, the ion beam was electrostatically raster scanned over a 0.5×1 cm² area. The total accumulated dose was precisely measured in-situ using a Faraday cup connected to a current integrator.

Structural changes in the irradiated graphene were characterized using a Renishaw InVia Reflex confocal Raman spectrometer. A 532 nm solid-state laser was used for excitation, focused onto the sample through a 100× objective lens with a numerical aperture of 0.9, resulting in a spot size of approximately 1 μm. To prevent any laser-induced heating or annealing of the defects, the incident power on the sample was kept below 1 mW. For each sample, spectra were acquired from at least ten different points to ensure and verify the homogeneity of the irradiation.

The gas sensing performance was evaluated in a custom-built, sealed stainless-steel chamber with a volume of approximately 50 cm³. The flow of gases into the chamber was precisely regulated using a set of calibrated mass flow controllers. The devices were mounted on a probe station inside the chamber, and electrical connections were made to external measurement instrumentation. All measurements were conducted at room temperature. The devices were first stabilized in a flow of dry air until a stable baseline resistance (R_0) was achieved. Subsequently, the target gas, 1000 ppm of hydrogen (H₂) balanced in dry air, was introduced into the chamber at the same flow rate of 10 sccm. The device resistance was continuously monitored by applying a constant DC bias voltage and measuring the current using the source meters (current: Keysight B2901A, voltage: Keysight 34970A). The sensor response (or sensitivity, S) is defined as the relative change in resistance: $S [\%] = (R_{gas} - R_0) / R_0 \times 100$, where R_{gas} is the resistance after a fixed exposure time of 600 seconds.

Table 1. Raman analysis results for deuteron (D^+) irradiated graphene samples, including the $I(D)/I(G)$ ratio, calculated the average inter-defect distance (L_D) and defect density (n_D)

Ions	Kinetic energy [keV]	Irradiation [ions/cm ²]	$I(D)/I(G)$	L_D [nm]	n_D [$\times 10^{11}$ defects/cm ²]
D^+	120	5×10^{13}	0.14	32.1	0.97
		1×10^{14}	0.15	31	1.04
		5×10^{14}	0.46	17.7	3.19
		1×10^{15}	0.62	15.3	4.27

STRUCTURAL CHARACTERIZATION OF ION-INDUCED DEFECTS

Raman spectroscopy provides a powerful non-destructive probe of the sp^2 carbon lattice, offering detailed information on defect structures.²⁹⁻³¹ The representative spectra of our samples are shown in Fig. 1. The pristine graphene exhibits a sharp G peak at $\sim 1595 \text{ cm}^{-1}$ and a symmetric $2D$ peak at $\sim 2700 \text{ cm}^{-1}$, with an intensity ratio $I(2D)/I(G) > 2$ and a negligible D peak $\sim 1352 \text{ cm}^{-1}$, all of which are characteristic of high-quality monolayer graphene.³² Upon deuteron irradiation, the D peak, which is activated by a double-resonant process involving defects, emerges and grows systematically in intensity with the ion dose. This allows for a quantitative analysis of the point-like defect density (n_D) using the well-established relation $n_D \approx 1/L_D^2$, where L_D is the average inter-defect distance calculated from the $I(D)/I(G)$ ratio.^{33,34} The calculated values for each dose are summarized in Table 1. The relationship between ion dose (Φ) and the resulting defect density is observed to be sub-linear, which can be fitted to a power-law function of the form $n_D \propto \Phi^\alpha$ with an exponent $\alpha < 1$. For D^+ ions, this yields an empirical relation: $I(D)/I(G) \approx 0.05 \cdot (\Phi/\Phi_0)^{0.54}$, with $\Phi_0 = 1 \times 10^{13} \text{ ions/cm}^2$. This sub-linear behavior arises from the defect overlap phenomenon at higher ion doses, the likelihood of ions hitting already damaged regions increases, reducing the efficiency of new defect formation.

As shown in Fig. 2. The stopping and range of ions in matter (SRIM) simulation were performed to confirm the physical mechanism of defect creation.^{35,36} The simulation shows that the 120 keV deuterons have a projected range of $\sim 1170 \text{ nm}$, meaning they come to rest deep within the Si substrate. This definitively rules out ion implantation as a source of chemical doping in the graphene. Furthermore, simulations of collision events confirm that the generation of atomic displacements, which requires transferring more than the displacement energy of carbon ($\sim 22 \text{ eV}$), begins at the very surface of the target, validating that the defects are created by kinetic energy transfer during the ion's transit through the graphene sheet.

NON-MONOTONIC CHEMIREISTIVITIVE RESPONSE

The dynamic sensor response to 1000 ppm of H₂ for devices with different irradiation doses is shown in Fig. 3. These curves demonstrate that both the initial response rate and the final sensitivity depend strongly on the defect concentration. As noted in the legend, R_0 increases monotonically with dose, from 419 Ω to 2290 Ω , which can be attributed to increased carrier scattering. The central result of this study is presented in Fig. 4, which plots S at $t=600$ s against n_D from Table 1. The data show a non-monotonic trend, where S increases with defect density up to an optimal point, beyond which it decreases. This Λ -shaped trend can be categorized into three distinct regimes. (i) Activation-Dominated Regime: At low defect densities ($n_D < 1.04 \times 10^{11}$ defects/cm²), the sensor sensitivity rises sharply and approximately linearly with n_D . The slope of this initial increase serves as a direct indicator of the effectiveness of the introduced defects as active adsorption sites. This suggests that the engineered defects contribute meaningfully to the adsorption of H₂ molecules and the associated charge transfer process. Since H₂ acts as a mild electron donor to p-type graphene, this interaction leads to hole recombination, thereby increasing the electrical resistance;³⁷ (ii) Optimal Point: The sensitivity reaches its peak value of 4.1 % at an optimal defect density of approximately 1.04×10^{11} defects/cm². At this point, the benefit of new active sites is balanced by the onset of performance degradation; (iii) Degradation-Dominated Regime: Beyond the optimal density, the sensor sensitivity gradually declines. In this regime, the adverse impact of defects on the charge transport properties of the graphene channel begins to outweigh the gains from additional active sites. The increasing number of scattering centers disrupts the sp² carbon network, limiting effective signal transmission from the adsorption sites to the electrodes and leading to diminished overall sensor response.^{7,38,39} This Λ -shaped trend can be categorized into three distinct regimes. (i) Activation-Dominated Regime: At low defect densities ($n_D < 1.04 \times 10^{11}$ defects/cm²), the sensor sensitivity rises sharply and approximately linearly with n_D . The slope of this initial increase serves as a direct indicator of the effectiveness of the introduced defects as active adsorption sites. This suggests that the engineered defects contribute meaningfully to the adsorption of H₂ molecules and the associated charge transfer process. Since H₂ acts as a mild electron donor to p-type graphene, this interaction leads to hole recombination, thereby increasing the electrical resistance;³⁷ (ii) Optimal Point: The sensitivity reaches its peak value of 4.1 % at an optimal defect density of approximately 1.04×10^{11} defects/cm². At this point, the benefit of new active sites is balanced by the onset of performance degradation; (iii) Degradation-Dominated Regime: Beyond the optimal density, the sensor sensitivity gradually declines. In this regime, the adverse impact of defects on the charge transport properties of the graphene channel begins to outweigh the gains from additional active sites. The increasing number of scattering centers disrupts the sp² carbon network, limiting effective signal transmission from the adsorption sites to the electrodes and leading to diminished overall sensor response.^{7,38,39}

PREDICTIVE MODEL FOR SENSOR SENSITIVITY

To quantitatively account for the observed Λ -shaped sensitivity behavior, we developed a physical model grounded in the fundamental mechanisms of chemiresistive sensing. The sensor response is approximated by the relative change in charge carrier concentration, expressed as $S \propto -\Delta n/n_0$, under the assumption that charge transfer dominates over mobility variations in this system.¹⁰ Here, n_0 is the initial carrier concentration and Δn is the change induced by gas exposure. Our model posits that the sensitivity is governed by the product of two competing physical effects. (i) Activation-Dominated Regime: The potential signal strength is proportional to the number of available active sites, represented by n_D ; (ii) Degradation-Dominated Regime: The efficiency of signal transduction decreases as the conductive lattice becomes increasingly disrupted. Assuming defects are introduced randomly and independently, their spatial distribution follows Poisson statistics. The probability that a critical signal pathway remains free of disruptive defects i.e., the zero-event probability decays exponentially with increasing defect density. By combining these effects, the sensitivity can be modeled as: $S(n_D) \approx C_n \cdot n_D \exp(-\gamma_n n_D)$, where C_n is the activation coefficient (or per-defect efficiency) and γ_n is the degradation coefficient (or lattice fragility). This expression is not merely an empirical fit; rather, it is derived from the physical principles that govern signal generation and transduction within the sensor, as well as from statistical theories describing charge transport in defect-rich materials. Given the limited dataset, a robust two-step fitting procedure is employed. First, γ_n is estimated from the experimentally observed peak position $n_{D,peak}$ using the relation $\gamma_n \approx 1/n_{D,peak}$. With γ_n fixed, C_n is determined by fitting the resulting single-parameter model to the full dataset. Using this approach, the extracted fitting parameters were determined to $C_n \approx 10.78 \times 10^{-11} \text{ \%} \cdot \text{cm}^2$ and $\gamma_n \approx 0.96 \times 10^{-11} \text{ cm}^2$. The fitted curve, represented by the dashed line in Fig. 4, exhibits good agreement with the experimental data. Despite the limited dataset, this simplified fitting method yields an intuitive and physically grounded approximation. Notably, the extracted parameters possess clear physical significance. C_n characterizes the intrinsic sensing efficiency of an individual defect site, while γ_n captures the structural fragility of the graphene lattice. With access to more extensive datasets, the parameter γ_n and C_n can be determined with greater statistical reliability using standard non-linear regression analysis. Accordingly, the relationship $n_{D,opt} = 1/\gamma_n$ serve as a practical design guideline, facilitating the a priori estimation of the optimal defect density required to maximize sensor performance.

CONCLUSION

This study proposes a quantitative and predictive model to better understand the non-monotonic sensitivity observed in defect-engineered graphene. By considering the balance between defect activation and lattice degradation, the model captures the characteristic Λ -shaped sensor response with good agreement to experimental trends. One of the main insights from this work is the identification of two physically interpretable parameters, per-defect efficiency (C_n) and the lattice fragility (γ_n), which help clarify the factors influencing sensor performance. These parameters offer a way to assess whether limitations arise from low defect activity or from a compromised charge transport pathway. In addition, the model provides a practical method for estimating the optimal defect density ($n_{D,opt} = 1/\gamma_n$), supporting

a more guided approach to sensor design compared to conventional trial-and-error methods. Although the current model simplifies the defect landscape and focuses primarily on signal magnitude, future studies can extend its applicability by incorporating a broader range of defect types and noise characteristics. Such efforts could contribute to more refined signal-to-noise optimization and improved device reliability.

Declarations

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Author Contributions

The manuscript was written through contributions of all authors. All authors have given approval to the final version of the manuscript. Sunmog Yeo, Junhyeok Seo, Young Jun Yoon, and Kibeom Kim contributed equally.

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Data availability statements

All data generated or analysed during this study are included in this published article.

Notes

Sunmog Yeo and Junhyeok Seo are co-first authors. Young Jun Yoon and Kibeom Kim are co-corresponding authors.

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Figures

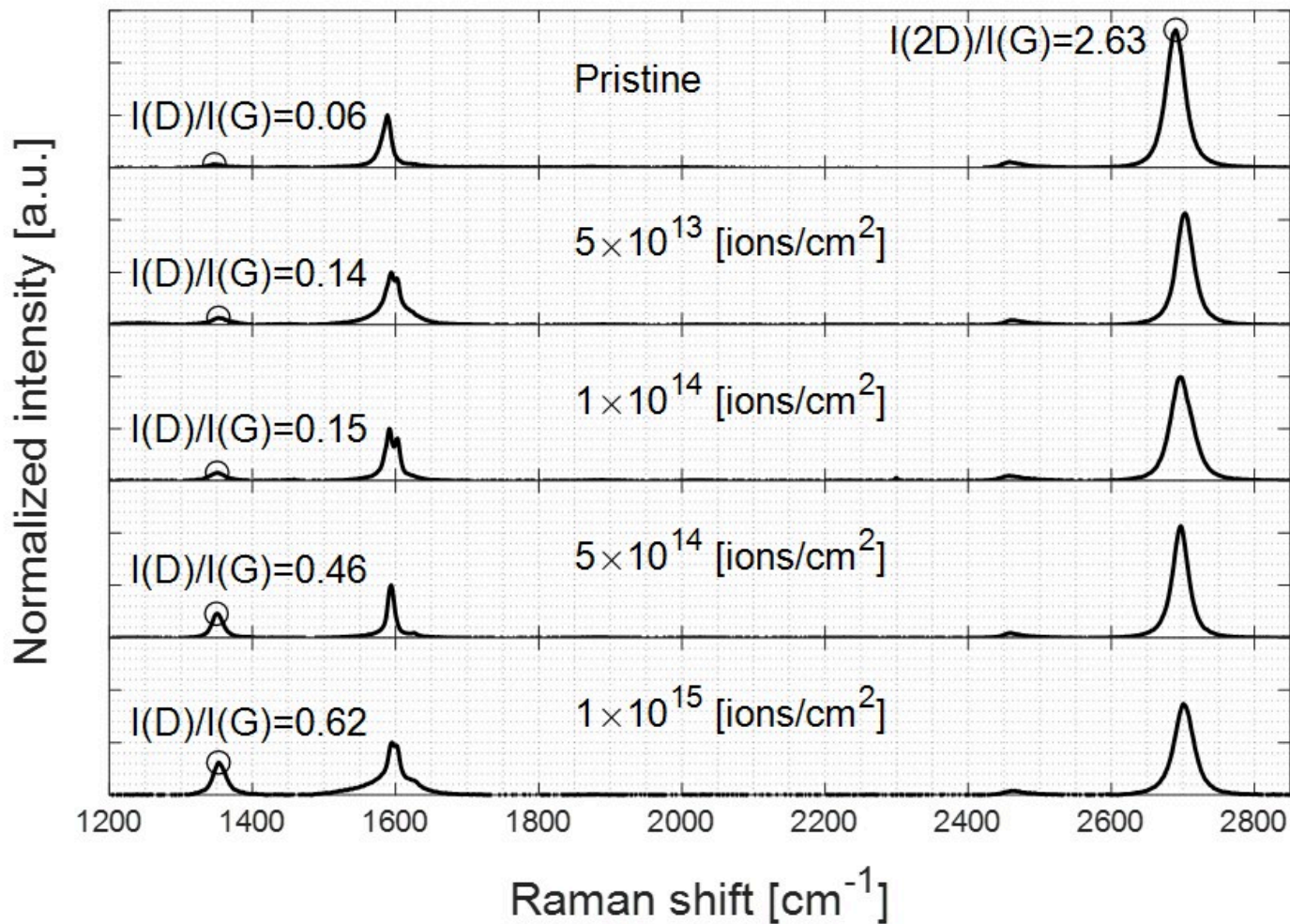
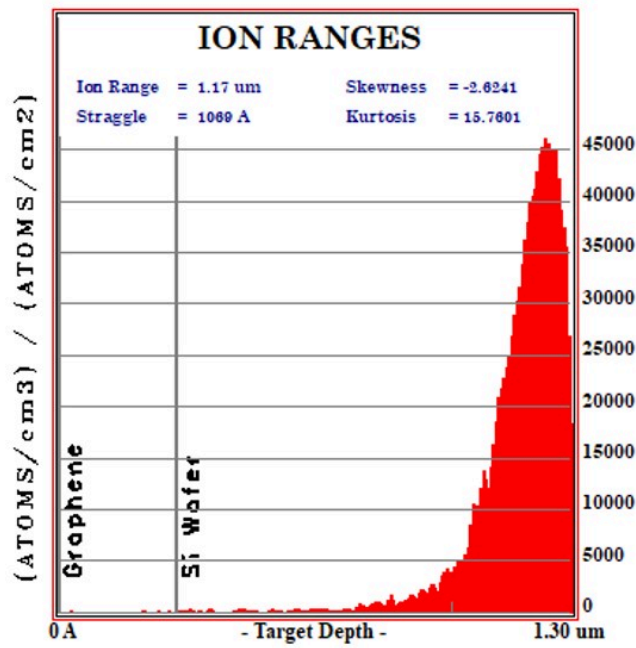
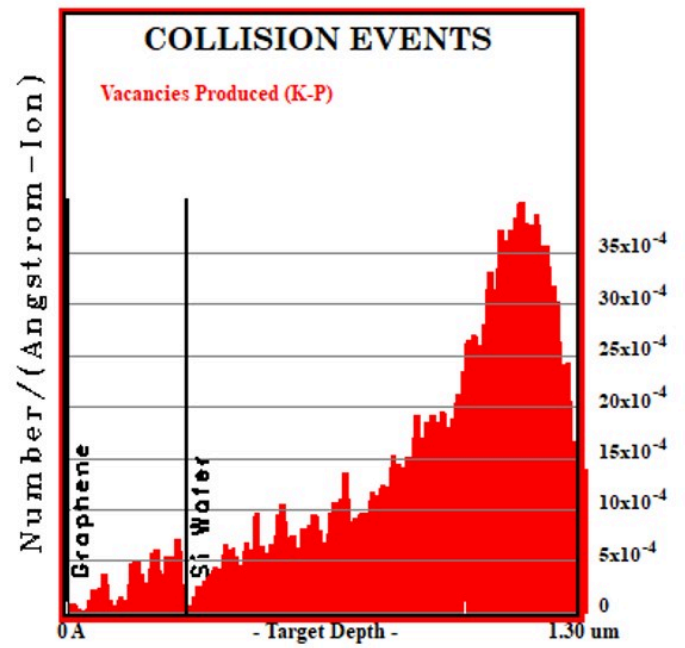


Figure 1

Raman spectra of a graphene sample irradiated with 120 keV D⁺ ions at increasing doses, from pristine (top) to 1×10^{15} ions/cm² (bottom). All spectra are normalized to the intensity of the graphitic G peak (~ 1582 cm⁻¹) for comparison. The increase in ion dose leads to a systematic growth of the defect-activated D peak (~ 1352 cm⁻¹), as quantified by the intensity ratio $I(D)/I(G)$.



(a)



(b)

Figure 2

SRIM simulation for 120 keV D^+ ions incident on the graphene/ SiO_2 /Si target. (a) The ion range distribution confirms that ions penetrate the graphene and stop deep within the Si wafer (~ 1170 nm). (b) The distribution of collision events shows that vacancy generation begins at the surface, within the graphene layer.

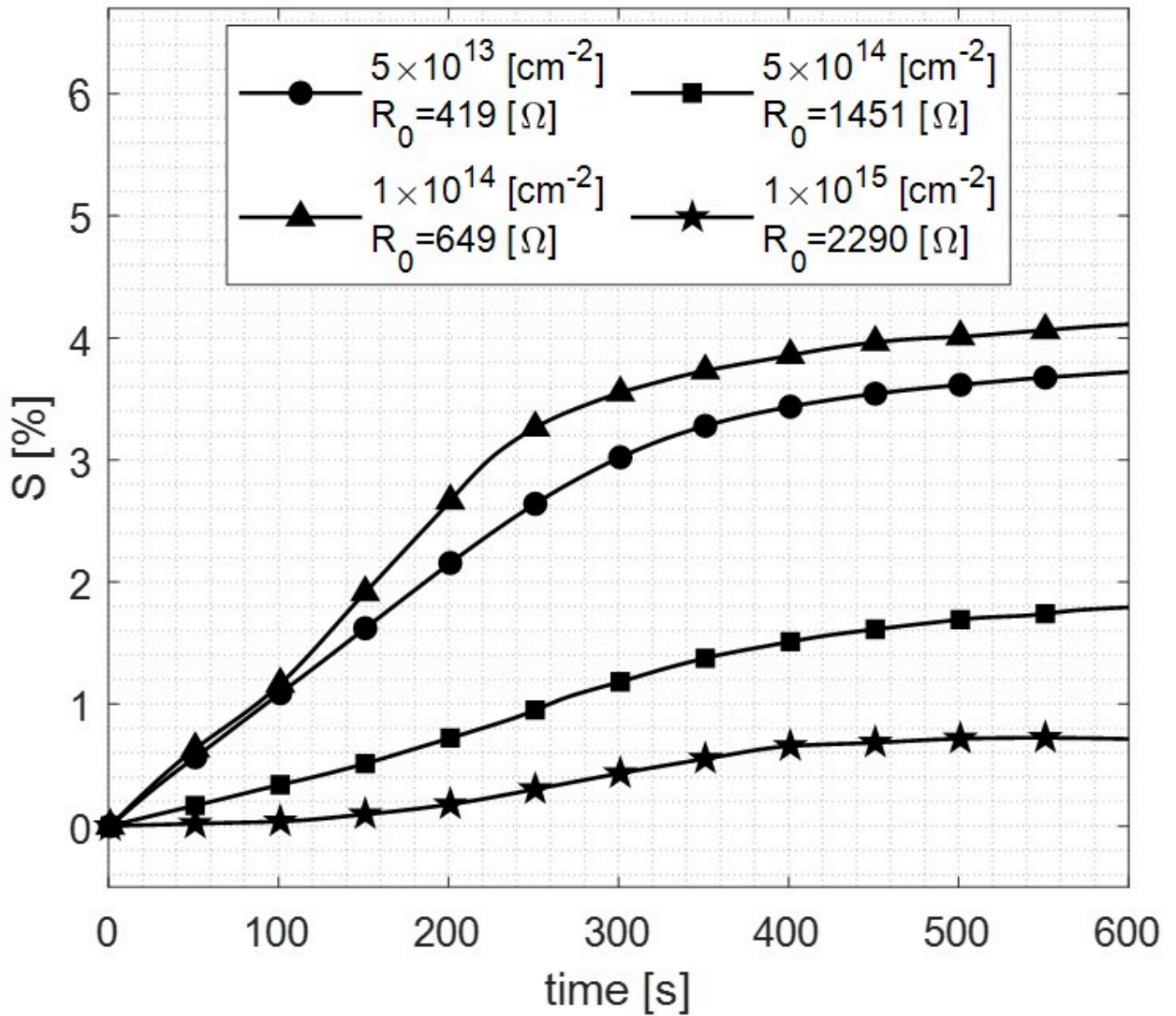


Figure 3

Real-time response of graphene sensors upon exposure to 1000 ppm H_2 gas. The y-axis represents the sensitivity (S), defined as $S(\%) = [(R_{gas} - R_0)/R_0] \times 100$, where R_{gas} is the resistance at a given time t , and R_0 is the initial baseline resistance. Each curve corresponds to a device fabricated with a different irradiation dose, with the specific dose and its measured R_0 value provided in the legend.

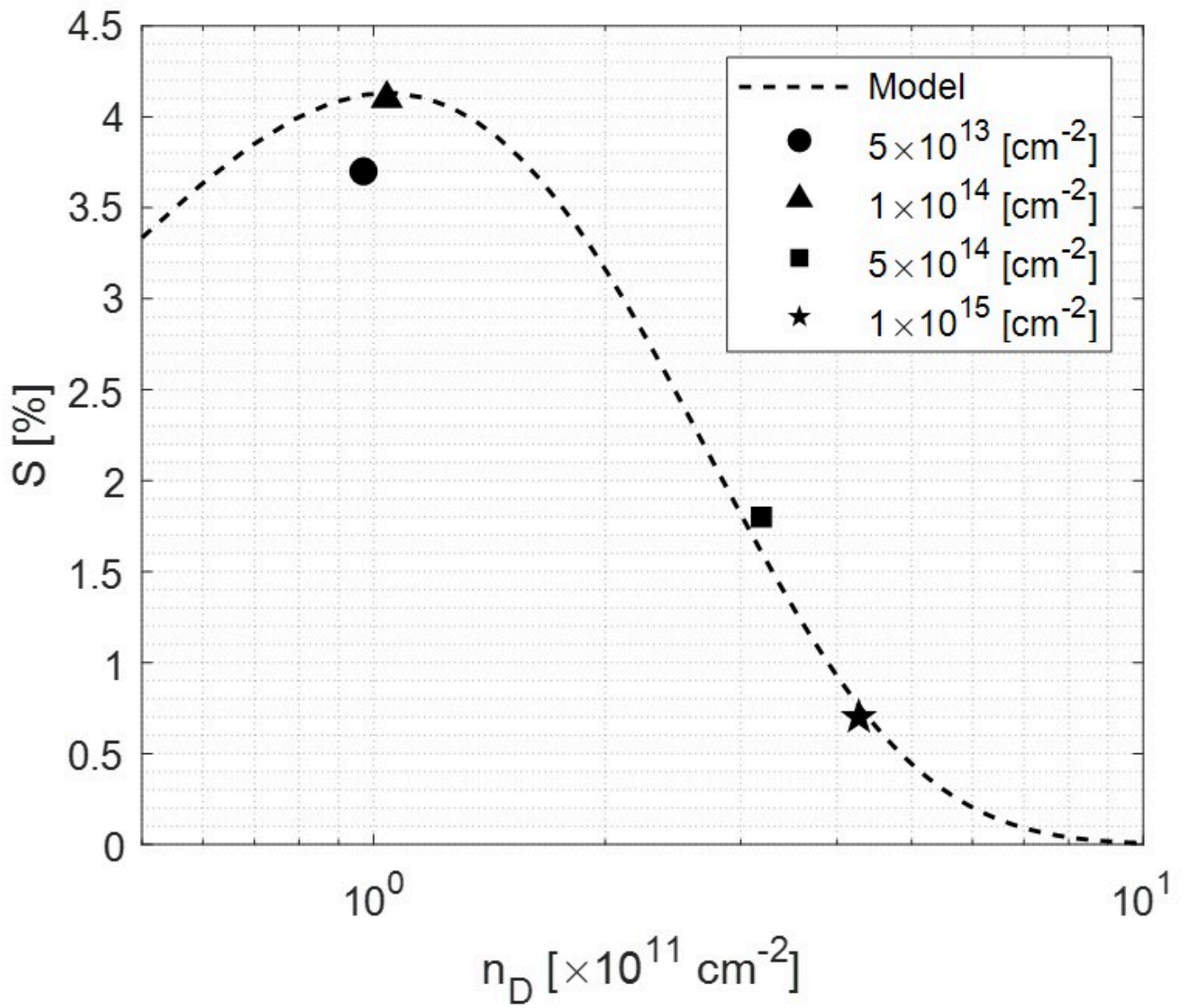


Figure 4

The non-monotonic relationship between sensor sensitivity (S) at $t = 600$ s and the calculated defect density (n_D). Experimental data are shown as markers, and the dashed line represents the fit from the proposed physical model.