Radiation tolerance test and damage of single-crystal CVD Diamond sensor under high fluence particles

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Abstract

Single-crystal chemical vapor deposition (CVD) diamond is a promising material for radiation detectors operating in extreme environments, owing to its outstanding radiation hardness. As nuclear and high-energy physics applications demand particle detectors that withstand higher radiation fluences, understanding the damage thresholds and degradation mechanisms of diamond-based detectors is essential for their practical operation. In this study, Synthetic single-crystal CVD diamond sensors were exposed to fast neutron irradiation at fluences up to 3.3$\times$1017 n/cm2, one of the highest test doses for evaluating radiation tolerance in diamond detectors. Modules exhibited stable signal output, retaining approximately 5% of their initial response after irradiation, confirming potential for application in future high-dose radiation environments. Fast neutron induced damage in the diamond crystals was characterized using photoluminescence and scanning electron microscopy. The dominant defects were identified as point defects including ⟨100⟩ self interstitials, vacancies, and lattice disorder. In addition, macroscopic defects on the crystal surface, including nanocavities and cracks, were observed with areal densities approaching 107/cm2. The impact of 100 MeV proton irradiation on diamond detector response was quantified by extracting a damage constant of k100MeVproton = (1.452 ± 0.006) × 10-18 cm2/(p · m) from a linear carrier drift degradation model. Moreover, the mean free path of carriers was found to exhibit saturation behavior beyond a fluence of 4$\times$1016 p/cm2 under 100 MeV proton irradiation. Monte Carlo together with molecular dynamics simulations were performed to assess irradiation induced defect production and its influence on carrier transport. The results indicate that saturation arises when local frenkel defect densities exceed 1018/cm3, at which defect interactions and clustering begin to dominate during irradiation. By considering saturation effects and defect-interaction corrections, we develop an enhanced carrier-drift degradation model that accurately captures detector response under high-dose irradiation. Furthermore, the simulation framework was applied to evaluate damage induced by protons and pions on diamond at various energies, yielding results that show better agreement with experimental data than conventional NIEL based estimates.

Full Text

Preamble

Radiation Tolerance Test and Damage of Single-Crystal CVD Diamond Sensor Under High Fluence Particles

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In this study, synthetic single-crystal CVD diamond sensors were exposed to fast neutron irradiation at fluences up to 3.3×10^{17} n/cm^2, one of the highest test doses for evaluating radiation tolerance in diamond detectors. Modules exhibited stable signal output, retaining approximately 5% of their initial response after irradiation, confirming potential for application in future high-dose radiation environments. Fast neutron induced damage in the diamond crystals was characterized using photoluminescence and scanning electron microscopy. The dominant defects were identified as point defects including 100 self-interstitials, vacancies, and lattice disorder. In addition, macroscopic defects on the crystal surface, including nanocavities and cracks, were observed with areal densities approaching 10^7 cm^{-2}. The impact of 100 MeV proton irradiation on diamond detector response was quantified by extracting a damage constant of k_{100} MeV proton = $(1.452 \pm 0.006) \times 10^{-18}$ cm^2/(p$\cdot$µm) from a linear carrier drift degradation model. Moreover, the mean free path of carriers was found to exhibit saturation behavior beyond a fluence of 4×10^{16} p/cm^2 under 100 MeV proton irradiation. Monte Carlo together with molecular dynamics simulations were performed to assess irradiation induced defect production and its influence on carrier transport. The results indicate that saturation arises when local Frenkel defect densities exceed 10^{18} cm^{-3}, at which defect interactions and clustering begin to dominate during irradiation. By considering saturation effects and defect-interaction corrections, we develop an enhanced carrier-drift degradation model that accurately captures detector response under high-dose irradiation. Furthermore, the simulation framework was applied to evaluate damage induced by protons and pions on diamond at various energies, yielding results that show better agreement with experimental data than conventional NIEL based estimates.

Keywords: Diamond detector, Radiation tolerance, Radiation damage, Defects simulation

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3.2. Radiation Induced Crystal Damage in Diamond

3.2.1. Luminescence Centers Associated with Radiation-Induced Defects

3.2.2. Irradiation-Induced Surface Morphology Characterization

3.3. Evaluation of the Radiation Hardness and Damage in Diamond with Damage Model

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Introduction

Over the past decades, particle physics has entered a transformative era, marked by unprecedented precision and discovery. The 2012 observation of the Higgs boson [?, ?] signaled a milestone, pushing experimental physics toward probing the Standard Model at increasingly finer scales while exploring potential new physics beyond it. These pursuits have driven accelerator and collider upgrades along both the luminosity and energy frontiers as well as the foundation of conceptual design for next-generation experiments [?], setting the stage for demanding operational conditions where detector radiation tolerance becomes critical [?]. In parallel, applications in nuclear fusion systems are approaching environments of comparable or even greater radiation severity [?], further amplifying the need for robust detection technologies.

Among candidate materials, synthetic single-crystal diamond has emerged as a leading contender for next-generation radiation detectors. It combines exceptional material properties, including high charge carrier mobilities larger than 3000 $\mathrm{cm}^2/(\mathrm{V}\cdot\mathrm{s})$ [?], a wide bandgap of 5.45 eV that enables excellent signal-to-noise performance, and superior thermal conductivity reaching 2000 $\mathrm{W}/(\mathrm{m}\cdot\mathrm{K})$ [?]. Most critically, its radiation hardness surpasses silicon by factors of three for low-energy incident particles and by more than a factor of ten at high energies [?, ?], which is attributed to its high displacement energy of up to 43.5 eV [?]. The advent of high-quality chemical vapor deposition (CVD) techniques has made large-area, defect-minimized single-crystal diamond (scCVD) substrates increasingly accessible [?, ?], making them a promising choice for applications in detectors subjected to high radiation levels. scCVD diamond detectors have been successfully employed in previous and ongoing experiments and facilities for beam monitoring and particle tracking, including the LHC, SuperKEKB, and the EAST tokamak system [?].

Despite these advances, the upper radiation dose limits that scCVD diamond detectors can tolerate without critical signal loss remain an open question. Prior studies, including work from RD42 [?, ?], have demonstrated the viability of diamond detectors under proton and neutron fluences approaching 10^{15} cm^{-2} to 10^{16} cm^{-2}. However, future environments are expected to push beyond these thresholds [?]. Moreover, irradiation sources often include mixed particle fields with broad energy spectra, making it essential to understand not only how damage accumulates but also how it impacts charge transport at the microscopic level and consequently affects detector performance. Detector performance degradation is commonly understood as a two-step process: irradiation introduces lattice defects, and these defects in turn act as traps or recombination centers, reducing carrier lifetimes and thus degrades charge collection efficiency and detector performance [?].

Although diamond is renowned for its exceptional radiation hardness, a detailed understanding of how irradiation-induced defects influence charge transport is still lacking. To address this gap, pioneering studies have carried out systematic irradiation experiments using protons and pions at multiple energies. These efforts produced quantitative data on signal degradation, which in turn enabled the development of simplified damage models [?, ?]. Such models have been applied to comparatively assess the effects of different particles and energy levels on diamond detector degradation [?]. Beyond these experimental insights, predictive understanding requires complementary modeling approaches. Two main strategies have been developed. The first relies on the concept of non-ionizing energy loss (NIEL) [?], where the energy deposited into the lattice leads to atomic displacements and crystal damage. The NIEL cross section provides a convenient and widely used metric for estimating radiation damage. The second strategy adopts a material-centric perspective, explicitly evaluating the number and types of defects generated in the lattice [?, ?]. This defect-informed methodology offers a more physically grounded framework for predicting detector degradation.

In this study, we present a comprehensive investigation into the radiation tolerance and damage mechanisms of scCVD diamond sensors exposed to high fluence fast neutron and protons. Four scCVD diamond sensors were fabricated and irradiated at nuclear facilities for a fast neutron radiation experiment. The cumulative neutron fluence achieved 3.3×10^{17} n/cm^2, one of the highest doses reported for single-crystal diamond to date. The sensors exhibited a sustained signal response, underscoring the potential of scCVD diamond as a viable candidate for mitigating the limited operational lifetime of silicon-based detectors, especially in the innermost layers of next-generation harsh radiation experiments. To elucidate the microscopic origins of radiation-induced performance degradation, the neutron irradiated diamonds were systematically characterized using photoluminescence spectroscopy and scanning electron microscopy. Damage was observed to generate atomic-scale point defects, crystalline lattice disorder, and macroscopic defects including voids and microcracks.

To assess the correlation between defect generation and transport degradation, we analyzed signal response data from scCVD diamond detectors previously irradiated with 100 MeV protons. Using a simplified carrier drift degradation model, we extracted a damage constant and normalized it to radiation damage of 24 GeV protons via established scaling relations [**?**], allowing direct comparison with results from other irradiation studies. This analysis contributes quantitative insight into an energy regime that has remained relatively unexplored. Notably, we observed that the carrier mean free path exhibited saturation behavior at high fluence levels, suggesting a shift in the dominant damage mechanisms. To interpret this effect, a combined simulation framework including Monte Carlo simulations and molecular dynamics modeling incorporating adiabatic recombination (arc-DPA) [**?**] were employed. The results indicate that when local defect densities exceed 10^{18} cm^{-3}, interactions among defects begin to dominate over isolated point defect formation, driving a transition toward saturation in performance loss. Based on this, we refined the traditional carrier drift degradation model to account for saturation effects at extreme doses. Finally, the combined simulation framework was applied to assess radiation damage from protons and pions across a range of energies. The predictions show closer agreement with experimental data than conventional NIEL-based estimates, highlighting the importance of defect-level modeling for accurate performance forecasting. Together, these results expand our understanding of diamond detector behavior under extreme radiation and provide actionable insights for their deployment in high-luminosity colliders, nuclear science such as fusion reactors, and space-based instruments.

2.1 Fabrication of DUT Modules

The single crystal diamond material was synthesized using a commercial 30 kW DC arc plasma jet chemical vapor deposition (CVD) system operated in gas recycling mode. On the substrates of commercial high pressure high temperature (HPHT) type-Ib (100) single-crystal diamond, a number of high quality large-sized single-crystal diamond plates were fabricated using the CVD homoepitaxial growth technique. In preparation for the fast neutron irradiation experiment, self-supporting synthetic scCVD diamond plates were separated from the substrates by laser cutting. Then, mechanical polishing and boiling with a combination of acids were also used in order to get rid of any potential contaminations and damaged layers on the surfaces of scCVD diamond. Following the cutting, polishing, and cleaning procedures, scCVD diamond plates have the final size with a surface area of 7.0 × 7.0 mm^2 and thickness around 300 µm, as depicted in Fig. 1 Figure 1: see original paper. Raman spectroscopy was carried out on the surfaces of the plates to examine the purity and perfection of the single-crystal diamond, the results [**?**] of strong first-order peak at 1332 cm^{-1} with a narrow full wave at half maximum (FWHM) of 2 cm^{-1} demonstrating the obtain of high quality synthetic scCVD diamond plates prepared for radiation detection sensors.

Planar Ti–W–Au electrodes were deposited on both surfaces of the scCVD diamond plates by magnetron sputtering, forming efficient metal–insulator–metal (MIM) detection sensors used as the Device Under Test (DUT) sensors, as illustrated in Fig. 1(a). The electrical properties of these sensors were assessed through I-V curve measurements, as depicted in Fig. 1(b). The curve reveals a minimal dark current of approximately 0.4 nA under a 500 V bias voltage with good linearity, indicating robust ohmic contact between the electrodes and the diamond. Subsequently, DUT sensors were incorporated into Rogers ceramic base high-frequency PCBs as modules, featuring planar electrodes connected to the readout electronics through gold wire bonding, as shown in Fig. 1(b). Long Kapton-insulated coaxial cables were utilized for electronic communication, facilitating a connection to the remote data acquisition (DAQ) system.

2.2 Radiation Tolerance Experiment

The fast neutron irradiation was undertaken at the IBR-2M reactor in the Laboratory of Neutron Physics, Joint Institute for Nuclear Research, Dubna, Russia. The experiment took place in the beamline specifically designed for neutron irradiation experiments [**?**]. The DUT modules were placed inside a container, which was positioned at one end of an extended conduit and located 30 centimeters away from the reactor moderator, to get high fluence of fast neutrons. Connected by long cables passing through a nuclear radiation shielded area, DUT modules were linked to the DAQ system. The DAQ system consists of a high voltage supply module, a voltage monitoring module, and a digital multimeter. The high-voltage supply module is responsible for providing and monitoring bias voltage up to 260 V on the DUT sensors during the experiment, supplied by the Keithley 6487 module. The multi-channel multi-meter Keithley 2700, equipped with the Keithley 7703 relay card, is used to read out and record DC current ionization detection signals triggered by fast neutrons, and transmit them to the computer. The schematic diagram of the entire process is shown in Fig.

2 Figure 2: see original paper. One HV line, not connected to the detector, was also read out to measure dark current and noise. The entire irradiation experiment accumulated for approximately 280 hours. During this period, the reactor' s operating power was maintained at around 2 MW, as shown in Fig. 2(b). The accumulated fast neutron irradiation fluence at the location of DUT modules reached 3.3×10^{17} n/cm^2, with a flux of 3.27×10^{11} n/cm^2/s.

3.3.3 Modification of the Simple Damage Model at High Radiation Dose

In Section 3.3.1, we applied a linear damage model to describe the degradation of single-crystal diamond under 100 MeV proton irradiation. The model captures the initial trend of radiation-induced performance loss and enables cross-comparison of damage constants. At very high fluences, however, the linear form diverges. In real crystals, atomic density and spatial volume are finite, placing an upper bound on the number of defects that can be generated. At the same time, the continued accumulation of irradiation-induced defects may ultimately drive structural transitions such as amorphization. Under such extreme conditions, the model is expected to break down. At high doses, irradiation data reveal that for 100 MeV protons, the previously observed linear relationship between carrier mean free path λ and particle fluence becomes invalid beyond approximately 4×10^{16} p/cm^2. As shown in Fig. 9 [Figure 9: see original paper], the mean free path begins to saturate, indicating that further damage accumulation no longer leads to proportional degradation in carrier transport.

In light of the observed deviation behavior, two types of nonlinear damage mechanisms are proposed. The first involves a saturation model of effective defect accumulation. Here, effective defects are defined as those that significantly contribute to the degradation of carrier lifetime, as opposed to the total population of structural disruptions within the crystal lattice. At high irradiation fluences, where the spatial density of energy deposition events increases significantly within localized regions of the crystal, certain areas may undergo early-stage amorphization or the formation of extended defect clusters, such as the defect clusters and sp^3-sp^2 phase transitions observed in irradiated diamond [**?**]. Subsequent energetic particles traversing heavily damaged and structurally disordered regions predominantly interact with existing amorphous networks or defect clusters that have already formed, depositing their energy within these disordered structures. In such amorphous domains, the electronic density of states extends into the gap, forming band tails and localized states due to disorder [**?**]. As a result, additional defects introduced into these regions tend to merge into existing localized states, contributing little further trapping or scattering. By contrast, when irradiation occurs in regions where the crystal lattice remains relatively ordered, newly generated defect levels act as efficient electrically active traps. These lattice defects are thus identified as the primary contributors of carrier transport degradation.

A saturation model of effective defect within a unit volume is proposed, we assume the existence of a saturation defect density n_{sat}, beyond which additional damage has a negligible effect on charge transport. When the local density of effective defects, n_{eff}, remains below n_{sat}, energy deposition by incident particles generates crystal damage that significantly degrades the carrier mean free path. Once n_{eff} exceeds n_{sat}, further energy deposition in these already disordered or cluster-rich regions is assumed to contribute little additional impact on drift behavior. Across the entire diamond crystal, the local effective defect density n_{eff} is expressed as a spatially averaged quantity N_{eff}. The evolution of the effective defect density is then described by:

$$N_{eff} = N_{sat} \cdot \left(1 - e^{-N(\phi)/N_{sat}}\right)$$

Using a combined Monte Carlo and molecular dynamics approach, the total defect number can be approximated by the relation N = N_{arc}( ) = k , as given by Equation 11. In the low-fluence regime, this expression naturally reduces to the previously linear damage model:

$$N \ll N_{sat} \implies N_{eff} \to N_{sat} \cdot \frac{N(\phi)}{N_{sat}} = N(\phi)$$

At excessively high fluence conditions, the effective defect concentration approaches a saturation limit:

$$N \gg N_{sat} \implies N_{eff} \to N_{sat} \cdot (1 - 0) = N_{sat}$$

By substituting N_{eff} into Equation 4, we obtain the damage expression under the effective defect model as $1/\lambda$ = k ·N_{eff} + $1/\lambda_0$. This approach provides a significantly improved description of the degradation behavior at high fluences, as seen by the blue curve in Fig. 13 [Figure 13: see original paper]. From the fitting, we extract the saturation defect density. For 100 MeV protons incident on diamond, this corresponds to approximately 2,036.8 defects within a 100 nm^3 unit volume as used in Geant4 simulations, which is equivalent to a bulk defect density of 2×10^{18} cm^{-3}. When the number of defects generated by radiation in a local region exceeds this threshold, saturation effects are expected to occur. If we estimate the saturation defect density as N_{sat} = N_{arc}($_0$), then the onset of saturation effects is expected to emerge at an equivalent fluence $_0$ of approximately 2.5 × 10^{16} p/cm^2.

Inspired by defect interactions and built on the estimation of overlapping irradiation-induced defects, we propose a second nonlinear damage mechanism, as detailed below. Molecular dynamics simulations were employed to estimate the spatial distribution of defects generated by PKAs in diamond. The resulting defect distributions for PKAs with varying recoil energies, obtained using the

LAMMPS package, are shown in Fig. 10 [Figure 10: see original paper]. As evident from the visualizations, higher-energy PKAs produce increasingly clustered defects. To quantify this behavior, we define a cutoff distance of 1.5 Å between carbon atoms and use it to statistically evaluate the size and number of defect clusters. The results are presented in Fig. 11 [Figure 11: see original paper]. These simulations reveal that a single PKA not only forms localized defect clusters but also establishes a characteristic spatial range over which its damage extends. As shown in Fig. 12 [Figure 12: see original paper], the cumulative number of clusters produced by a PKA increases with recoil energy, and dislocation defects are also observed in the simulations. These findings indicate that each PKA generates a distinct damage volume comprising energy-dependent cluster regions and extended defects.

At high fluences, the probability that a new PKA' s damage volume overlaps with pre-existing damage regions rises. We hypothesize that within these overlapping regions, interactions between clusters dominate over the formation of isolated point defects. Furthermore, we assume that such overlapping volumes contribute minimally to additional carrier degradation. Consequently, only non-overlapping PKA damage regions are considered effective in reducing carrier transport. We define the effective damage volume as V_{eff}( ) = V_{tot}( ) – V_{overlap}( ), and introduce a fluence-dependent damage reduction factor:

$$k_{overlap}(\phi) = \frac{V_{tot} - V_{overlap}}{V_{tot}}$$

In the Monte Carlo simulations, the spatial positions of individual PKAs were recorded. The total damage volume, V_{tot}, was obtained by summing the effective defect volumes produced by each PKA over the entire irradiation period. Simultaneously, the overlapping volume, V_{overlap}, was determined by calculating the intersection between newly generated PKA defect volumes and those already existing. For each PKA, the associated damage volume was derived by fitting molecular dynamics simulation results (Fig. 10) for various recoil energies. Finally, by replacing the constant damage parameter in Eq. (4) with the fluence-dependent damage reduction factor at each fluence, we obtained the carrier mean free path as a function of fluence according to the second nonlinear damage mechanism, as shown by the orange curve in Fig. 13 [Figure 13: see original paper]. This modified model, incorporating defect overlap, reproduces the damage behavior observed at high fluence.

To further illustrate this effect, we consider a diamond crystal with a volume of 300 nm^3. At different proton fluences, denoted as = 1.2 × 10^{16}, = 2.4 × 10^{16}, = 3.6 × 10^{16}, = 4.8 × 10^{16}, = 6.0 × 10^{16}, = 7.2 × 10^{16}, = 8.4 × 10^{16}, = 9.6 × 10^{16}, = 1.08 × 10^{17}, = 1.2 × 10^{17}, the number of PKAs and their corresponding spatial damage ranges are visualized in Fig. 14 [Figure 14: see original paper]. In the figure, the circular mappings represent the extent of defect-affected regions, while different colors correspond to the defect volumes generated by different PKAs. Lighter colors indicate larger affected volumes. It is evident that as the fluence exceeds around 3 × 10^{16} p/cm^2 to 4 × 10^{16} p/cm^2, the overlapping regions between PKA-induced damage volumes increase significantly, leaving less undisturbed crystal volume for the formation of isolated point defects. From the perspective that each PKA generates multiple defect clusters, the increasing dominance of defect–defect interactions becomes apparent at high fluences.

Through the effective defect saturation model and the defect interaction model, this study establishes a connection between irradiation-induced defect evolution and carrier transport degradation in diamond, providing a phenomenological framework that captures the nonlinear behavior of diamond detectors under high radiation fluence. The first model assumes a saturation of electrically active defects capable of trapping carriers, while the second considers the spatial overlap and interaction among newly generated and pre-existing defects. Together, they account for the observed deviation from linearity in detector response at high doses. Both frameworks converge on a consistent interpretation that under intense irradiation the diamond lattice undergoes a structural transformation from a regime dominated by isolated point defects to one governed by defect clusters and locally amorphous configurations. This structural transition in radiation damage modifies the distribution of density of states within the band structure, subsequently changing the effective carrier trapping cross section and driving the detector response from linear to nonlinear behavior in CCD and other electrical characteristics. Monte Carlo and molecular dynamics simulations indicate that this transition begins to emerge when the defect density approaches 10^{18} cm^{-3}, corresponding to an equivalent neutron fluence of 10^{16} n/cm^3, where linear scaling with fluence no longer holds. In this regime, comprehensive understanding of these nonlinear processes will require integrated first principles calculations and advanced experimental probes capable of resolving the atomic-scale mechanisms underlying defect clustering and amorphization in diamond.

Conclusion

This study demonstrates that single-crystal CVD diamond sensors retain functional signal response under exceptionally high radiation fluence, maintaining approximately 5% of their initial output after fast neutron irradiation up to 3.3 × 10^{17} n/cm^2 among the highest levels tested to date. These results confirm the feasibility of applying such sensors in extreme radiation environments.

Spectroscopic and electron microscopy analyses revealed that both bulk and surface defects induced by irradiation, such as self-interstitials, vacancies, and nanoscale surface cracks play a central role in the observed degradation of detector performance. Following the linear carrier-drift degradation framework, we experimentally extracted the quantitative damage constant for 100 MeV protons based on low fluences experimental data as k_{100} MeV = 1.452(6) × 10^{-18} $cm^2/(p \cdot \mu m)$, providing essential reference data for radiation damage assessment in diamond under medium-energy proton irradiation conditions.

To explore the underlying mechanisms of damage saturation, we performed multiscale simulations that couple Monte Carlo particle transport with molecular dynamics modeling. This combined approach yields damage estimates that align more closely with experimental observations than conventional NIEL predictions, offering a new perspective for studying radiation damage in diamond detectors. Furthermore, the framework provides a phenomenological means to investigate the observed saturation in carrier transport at high doses and to refine nonlinear degradation models. At high fluences, interactions between defects may become the dominant mechanism of lattice modification, gradually replacing isolated point defect formation. This transition leads to the emergence of a local effective saturation defect density, beyond which additional damage has a diminishing effect on the carrier drift length.

In summary, these findings establish a fundamental understanding of radiation-induced damage in diamond at high fluence, and offer practical guidance for the design and deployment of diamond-based detectors in future high-radiation particle physics experiments and advanced nuclear technologies.

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Author Contributions

All authors contributed to the work. Jialiang Zhang, Shuo Li, Guojun Yu, and Zifeng Xu performed characterization experiments, data analysis, simulations, and manuscript writing. Yilun Wang and Shuxian Liu contributed to model analysis. Lifu Hei and Fanxiu Lv provided materials. Ming Qi contributed to experimental design, planning, conducted irradiation experiments, and provided funding support. Ming Qi and Lei Zhang were responsible for manuscript revision and supervision. The first draft of the manuscript was written by Jialiang Zhang, and all authors commented on previous versions. All authors have read and approved the final manuscript.

Data Availability

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

Declaration of Interests

The authors declare that they have no known competing interests or personal relationships that could have appeared to influence the work reported in this paper.

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