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Defect and Interface Engineering in ZnO/g-C_3N_4 Heterostructures for Solar Photocatalysis: A Solid-State Physics Perspective

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Abstract: *ZnO/g-C_3N_4 heterostructures provide a promising platform for improving solar photocatalysis through controlled defect chemistry and interfacial electronic interactions. However, photocatalytic enhancement is frequently interpreted using simplified band diagrams without adequately considering defect states, work-function mismatch, band bending, interfacial charge redistribution, and surface reaction kinetics. This paper presents a solid-state-physics perspective on defect and interface engineering in ZnO/g-C_3N_4 heterostructures, emphasizing the relationship between band-edge energetics, electronic structure, charge-carrier dynamics, and surface redox processes. The charge-transfer characteristics of Type-II, direct Z-scheme, and S-scheme configurations are critically compared, with particular emphasis on the experimental evidence required for distinguishing these mechanisms. A Band–Interface–Carrier–Reaction (BICR) framework is proposed as a systematic approach for correlating material structure, interfacial properties, carrier behaviour, and photocatalytic reactions. The framework integrates structural and optical characterization with band-structure, surface-potential, electrochemical, and reactive-species analyses. Rather than introducing unsupported experimental values, the study establishes experimentally testable criteria for mechanism validation and rational heterostructure design. This perspective provides a physics-based foundation for developing ZnO/g-C_3N_4 and related 2D semiconductor systems for solar-driven environmental remediation.*

Key Words: *ZnO/g-C_3N_4 heterostructures, Defect engineering, Interface engineering, S-scheme charge transfer, Charge-carrier dynamics, Solar photocatalysis*

I. INTRODUCTION

Solar-driven semiconductor photocatalysis is an interdisciplinary field in which solid-state electronic structure determines the initial photoexcitation process while surface chemistry determines the subsequent redox reactions. When a semiconductor absorbs photons with energy equal to or greater than its band gap, electrons are excited from the valence band to the conduction band, leaving photogenerated holes in the valence band. The useful fraction of these carriers must then migrate toward reactive surface sites before recombination. Thus, photocatalytic activity is controlled by a coupled sequence of optical absorption, carrier generation, transport, separation, trapping and surface reaction.

Pristine semiconductor photocatalysts frequently face a trade-off between broad light absorption and sufficiently strong redox potentials. In addition, Defects can introduce localized electronic states that either promote charge trapping and interfacial transfer or act as recombination centers, depending on their nature, concentration, energetic position, and spatial distribution. Heterostructure formation is therefore widely investigated as a route to modify the interfacial electronic structure and provide a directional pathway for photogenerated carriers [1–4].

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Graphitic carbon nitride is a visible-light-responsive, metal-free semiconductor with an optical band gap commonly reported around 2.7–2.8 eV, although the measured value depends on synthesis, morphology, defects and analysis method [5–7]. ZnO is a wide-band-gap semiconductor with high electron mobility and established photocatalytic functionality. Coupling ZnO with g-C_3N_4 can alter optical absorption and interfacial charge transport and has been explored using both Z-scheme and S-scheme interpretations [8–11].

Recent literature makes the mechanistic issue particularly important. A 2024 study reported a ZnO/g-C_3N_4 system interpreted as a direct Z-scheme for dye wastewater treatment, while a 2025 study combined experimental work with density-functional-theory and radical-scavenging analysis to support an S-scheme interpretation for g-C_3N_4/ZnO [9,10]. These studies demonstrate that the same material pair can be discussed using different charge-transfer models depending on synthesis, interface chemistry and evidence.

The purpose of the present paper is therefore not to claim a new ZnO/g-C_3N_4 material or experimentally establish a specific charge-transfer mechanism without direct experimental verification. Instead, this study develops a solid-state-physics-oriented framework for addressing a more fundamental question: how can band-edge energetics, defect states, interface properties, and charge-carrier dynamics be systematically correlated with the observed photocatalytic response? To address this question, a Band–Interface–Carrier–Reaction (BICR) framework is proposed to connect semiconductor electronic structure and defect chemistry with interfacial charge redistribution, carrier behavior, and surface redox processes. Particular emphasis is placed on experimentally testable criteria for distinguishing Type-II, direct Z-scheme, and S-scheme mechanisms and on establishing a rational basis for defect and interface engineering in ZnO/g-C_3N_4 and related 2D semiconductor heterostructures.

II. SOLID-STATE PHYSICS OF SEMICONDUCTOR PHOTOCATALYSIS

2.1 Photon Absorption And Carrier Generation:

For an idealized semiconductor transition, the minimum photon energy required for excitation is related to the band gap by:

$$E_{g)} = E(C) - E(V)$$

The corresponding threshold wavelength can be approximated by $\lambda g \approx 1240/Eg$, where λg is in nm and Eg is in eV. This relationship provides a simple first estimate of the spectral region accessible to a photocatalyst. It does not, however, determine photocatalytic efficiency by itself because absorption, quantum yield and charge-transfer kinetics are separate quantities.

2.2 Charge Generation And Recombination:

Following photoexcitation, photogenerated electrons and holes undergo competing processes, including transport, trapping, interfacial transfer, and recombination. A simplified carrier-balance equation can be written as:

$$\frac{dN}{dt} = G - (k_r + k_t)N$$

where N is the carrier population, G is the generation rate, k_r is the effective recombination rate constant, and k_t represents productive trapping or charge-transfer processes. In heterostructures, the objective is to promote spatially favorable carrier separation and interfacial transfer while suppressing recombination.

2.3 Defect States:

Defects introduce localized electronic states within or near the band gap and can substantially modify carrier dynamics and interfacial electronic structure. Depending on their type, concentration, energetic position, and spatial distribution, defects may act as beneficial charge-trapping sites or as recombination centers. Controlled defect engineering can therefore improve photocatalytic performance, whereas excessive defect density may increase carrier loss [6,12,13].

In ZnO/g-C_3N_4 heterostructures, defects such as oxygen vacancies in ZnO and carbon- or nitrogen-related defects in g-C_3N_4 may influence local charge density, band-edge positions, and interfacial charge redistribution. However, the presence and role of specific defects should be established using appropriate experimental evidence rather than inferred solely from enhanced photocatalytic activity.

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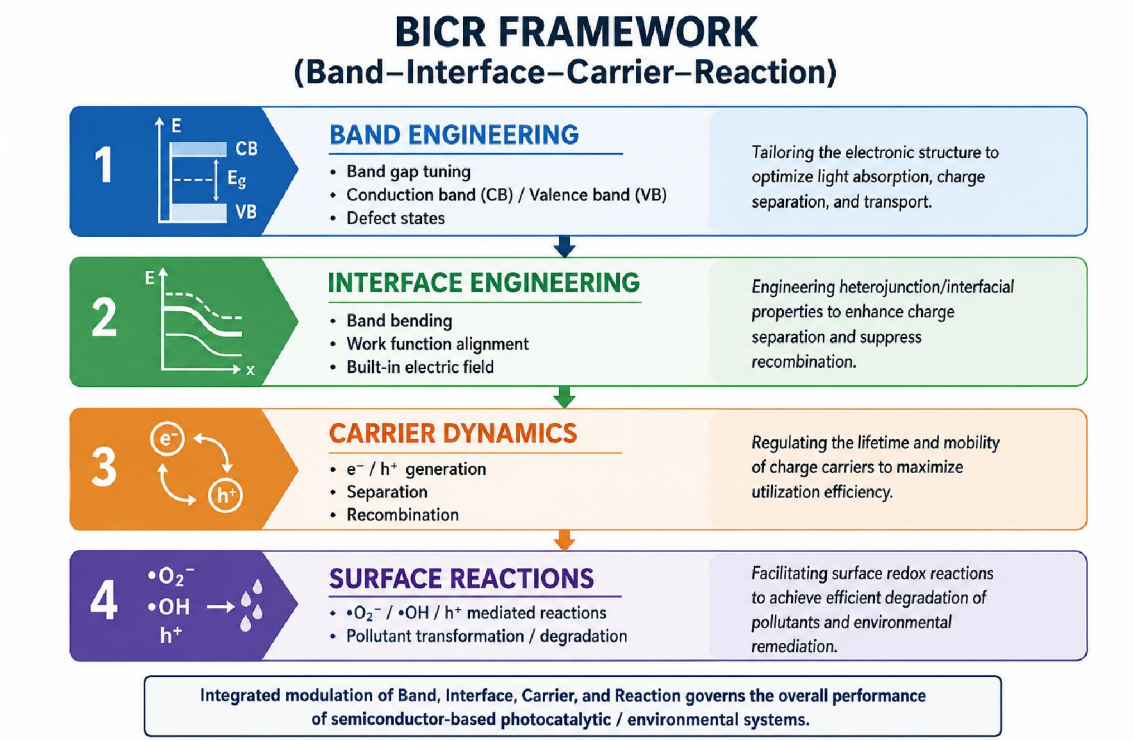


Figure 1. Proposed Band–Interface–Carrier–Reaction (BICR) framework for linking solid-state properties to photocatalytic performance.

III. ZNO/G-C_3N_4 AS A MODEL HETEROSTRUCTURE:

ZnO/g-C_3N_4 is a useful model system for a physics-centered study because it combines an inorganic metal-oxide semiconductor with a polymeric, conjugated carbon nitride semiconductor. g-C_3N_4 is generally described as a visible-light-responsive material with a band gap near 2.7 eV, while ZnO has a substantially wider band gap. The exact band-edge positions of either component are synthesis-dependent and should therefore be measured for the specific samples under investigation rather than imported uncritically from literature [5–7].

The formation of an intimate interface can change the electronic structure relative to a physical mixture. Differences in work function can drive charge redistribution after contact, producing an interfacial electric field and possible band bending. Such effects are central to the S-scheme interpretation. At the same time, direct Z-scheme descriptions may be appropriate for interfaces where selective carrier recombination retains the high-energy electrons and holes required for strong redox reactions. Distinguishing these models requires converging evidence [2–4].

IV. BAND ALIGNMENT AND HETEROJUNCTION MECHANISMS:

4.1 Type-II heterojunction

In a conventional staggered Type-II arrangement, photoexcited electrons tend to accumulate at the lower conduction-band side and holes at the higher valence-band side. This spatial separation can reduce recombination but may also weaken the reduction and oxidation potentials of the surviving carriers. Therefore, a lower photoluminescence intensity or higher photocurrent alone does not establish Type-II behavior.

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4.2 Direct Z-scheme

A direct Z-scheme aims to preserve strongly reducing conduction-band electrons on one component and strongly oxidizing valence-band holes on the other, while the less useful carriers recombine across the interface. This model can explain strong redox activity without requiring a conventional Type-II carrier pathway [4,8,9].

4.3 S-scheme

The S-scheme model introduces an explicit solid-state picture involving work-function differences, Fermi-level equilibration, charge redistribution, band bending and an internal electric field. In the dark state, charge transfer toward Fermi-level equilibrium can create an interfacial potential landscape. Under illumination, the resulting field and band bending favor selective retention of the strongly reducing electrons and strongly oxidizing holes [2,3].

Because Type-II and S-scheme diagrams can appear superficially similar, mechanism assignment should not be based on band-edge positions alone. Reviews published in 2024 specifically emphasize the need for complementary measurements when distinguishing Type-II from S-scheme behavior [2–4].

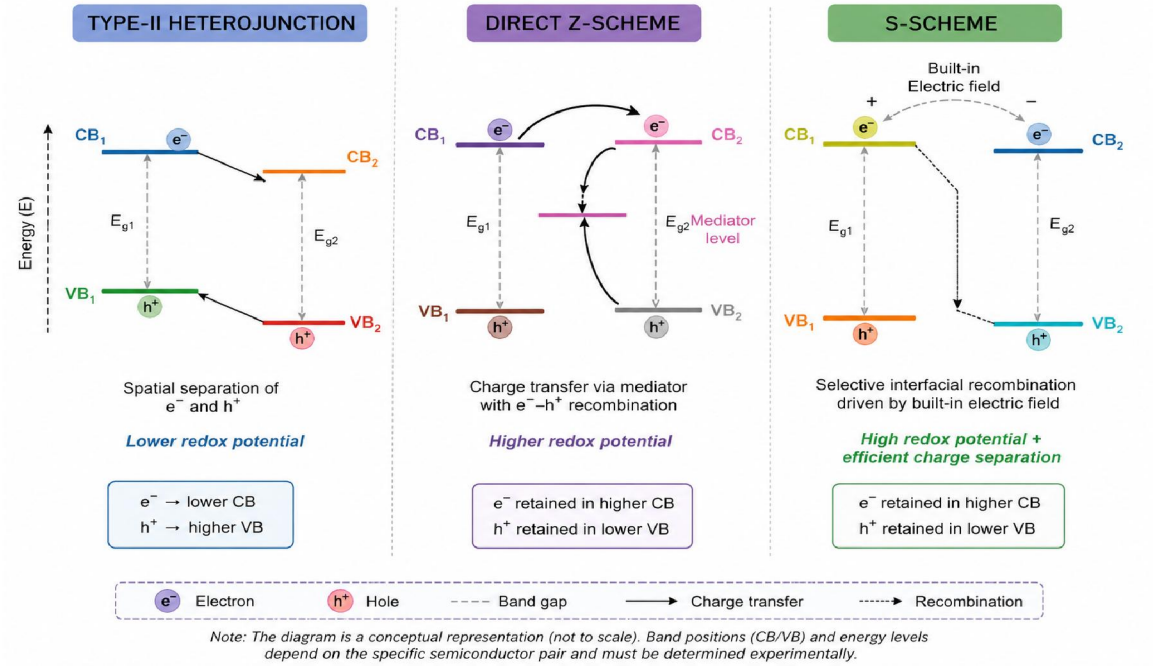


Figure 2. Conceptual comparison of Type-II, direct Z-scheme and S-scheme charge-transfer models. Band positions are schematic and not to scale.

V. EVIDENCE-BASED MECHANISM VALIDATION:

A major methodological issue in heterojunction photocatalysis is the tendency to infer a complete mechanism from two or three measurements. A stronger approach is to construct an evidence chain in which each technique addresses a different physical question.

Property	Technique	Primary Information	Mechanistic Value
Crystal phase	XRD	Phase, crystallinity, lattice changes	Confirms component formation; not sufficient for charge-transfer assignment
Morphology/interface	SEM/TEM/HRTEM	Particle size, contact, lattice interface	Supports intimate heterointerface

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Optical absorption	UV–Vis DRS	Absorption edge, apparent Eg	Assesses light harvesting
Carrier recombination	PL/TRPL	Emission and carrier lifetime	Supports recombination analysis
Band positions	UPS/XPS/Mott–Schottky	VB/CB and semiconductor type	Constrains possible pathways
Surface potential	KPFM/SPV	Work function, potential changes	Important for S-scheme/interface-field analysis
Charge transport	EIS/photocurrent	Resistance and photoresponse	Supports improved transport/separation
Reactive species	EPR/scavengers	$\bullet O_2^-$, •OH, h^+ etc.	Connects carriers to surface chemistry
Transformation	HPLC/LC-MS/TOC	Products and mineralization	Distinguishes removal from actual remediation

Mechanistic assignment becomes reliable only when independent measurements converge on the same carrier-transfer pathway. For example, an S-scheme claim would be more credible when band energetics, work-function/surface-potential information, light-induced charge redistribution, reactive-species data and electrochemical response point toward the same carrier pathway. No single measurement should be treated as definitive.

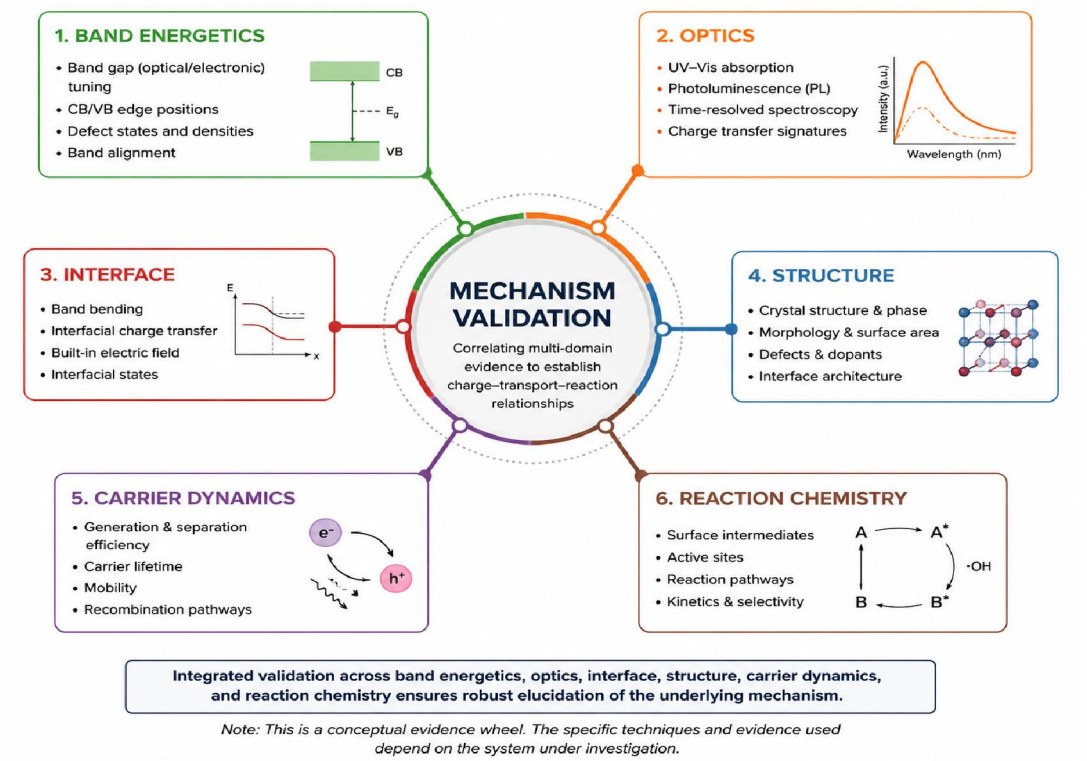


Figure 3. Evidence wheel for mechanism validation. The figure is conceptual; no artificial percentage weighting is assigned.

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VI. DEFECT ENGINEERING: OPPORTUNITY AND RISK:

Defect engineering should be treated as an electronic-structure problem rather than merely a synthesis modification. A defect can introduce localized states inside the forbidden gap, alter carrier trapping, modify adsorption energy or change the interfacial electric field. The same defect may therefore enhance one step while suppressing another. Recent reviews on g-C_3N_4 emphasize this dual role, noting that defects can improve light absorption and carrier separation but can also create recombination centers when uncontrolled [6,12,13].

For ZnO/g-C_3N_4, oxygen vacancies are particularly relevant because they can modify the electronic structure of ZnO and influence surface adsorption. Recent experimental studies have reported oxygen-vacancy-rich ZnO/g-C_3N_4 systems and linked these defects with enhanced photocatalytic response [14,15]. However, the concentration, spatial distribution and chemical state of vacancies should be established by appropriate spectroscopy rather than inferred from activity alone.

A physics-guided optimization strategy should therefore search for a defect concentration that maximizes productive carrier separation without increasing trap-assisted recombination. This suggests an experimentally testable non-monotonic relationship between defect density and photocatalytic performance.

VII. PROPOSED EXPERIMENTAL VALIDATION STRATEGY:

The present manuscript is developed as a perspective/review framework and does not report new experimental measurements. Instead, it proposes a systematic experimental workflow for future validation of the electronic structure, interfacial charge-transfer behaviour, and photocatalytic mechanism of ZnO/g-C_3N_4 heterostructures. The proposed workflow integrates complementary structural, optical, electronic, electrochemical, and reactive-species analyses to establish an evidence-based framework for mechanism verification.

Experimental validation should begin with structural and compositional characterization to confirm the formation and interfacial coupling of the ZnO/g-C_3N_4 heterostructure. Optical and electronic-structure analyses should then be employed to determine the optical band gap, band-edge positions, and interfacial electronic characteristics. Charge-separation and charge-transport behaviour can subsequently be assessed using steady-state and time-resolved photoluminescence, transient photocurrent measurements, and electrochemical impedance spectroscopy. Finally, radical-scavenging experiments and/or electron paramagnetic resonance spectroscopy can be used to identify the dominant reactive species involved in the photocatalytic process. Correlating these independent lines of evidence with photocatalytic activity, kinetics, and stability would provide a robust basis for evaluating the proposed interfacial charge-transfer pathway.

This evidence-based validation strategy is intended to distinguish experimentally supported charge-transfer mechanisms from pathways inferred solely from band alignment or photocatalytic performance. Such an integrated approach can improve the reliability and reproducibility of mechanistic interpretations of ZnO/g-C_3N_4 heterostructures

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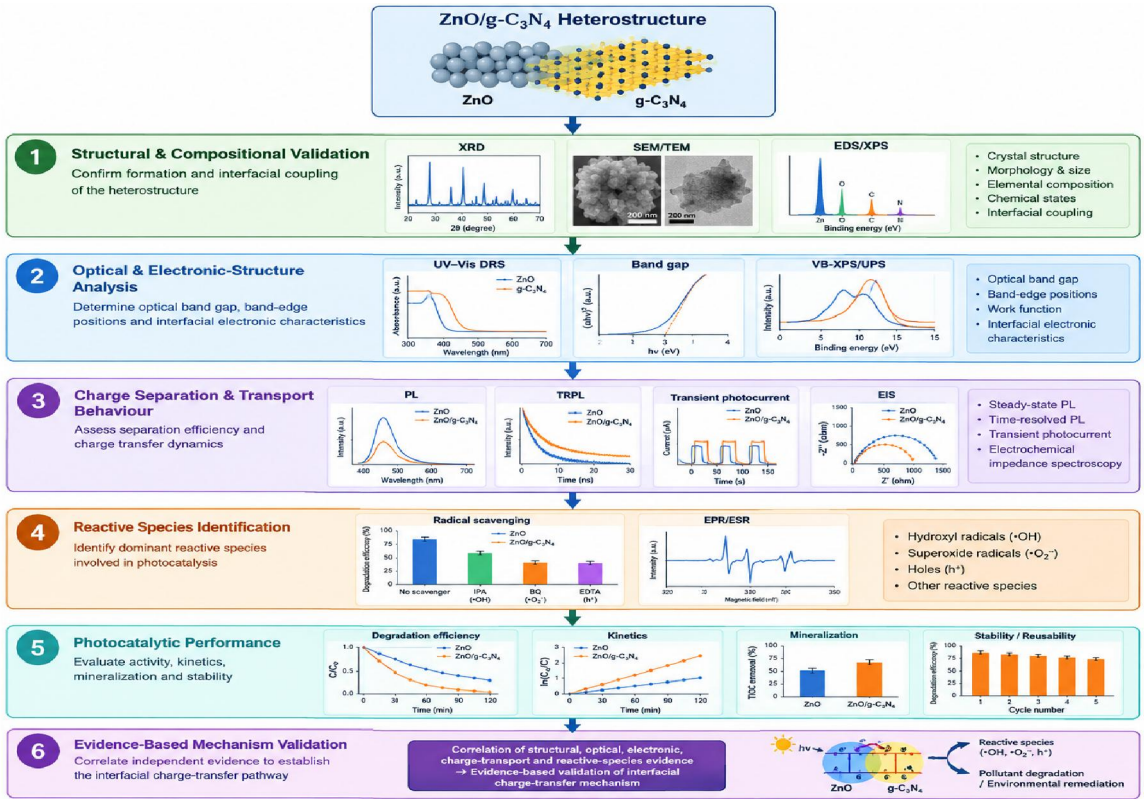


Figure 4. Proposed experimental validation workflow for a ZnO/g-C_3N_4 heterostructure.

VIII. PHOTOCATALYTIC WATER-REMEDIATION EVALUATION:

Pollutant removal is often reported as a percentage, but a robust materials assessment should include kinetics, mineralization, stability and reaction products. The apparent removal efficiency can be expressed as:

$$Removal\ (\%) = [(C_0 - C□)/C_0] \times 100$$

For a pseudo-first-order approximation under suitable experimental conditions:

$$ln(C_0/C□) = k_a□□\ t$$

Evaluation level	Recommended measurement	Why it matters
Removal	UV–Vis or chromatography	Measures disappearance of parent compound
Kinetics	Time-resolved concentration	Allows comparison of rates under defined conditions
Mineralization	TOC	Tests whether organic carbon is substantially mineralized
Transformation products	HPLC/LC-MS	Identifies possible intermediates
Active species	EPR/scavengers	Connects carrier chemistry to reactions
Stability	Multiple cycles + XRD/XPS	Tests structural/chemical durability
Energy relevance	Irradiance, reactor geometry, energy input	Improves comparability and scale-up assessment

where C_0 is the initial concentration, C□ is the concentration at time t, and k_a□□ is the apparent rate constant. The model should only be used after checking that the experimental data are consistent with the assumed kinetics.

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IX. STRUCTURE–PROPERTY–PERFORMANCE CORRELATION

The central hypothesis of this perspective is that photocatalytic performance should be interpreted through a chain of causality rather than through isolated measurements:

Defect/interface structure → electronic structure → carrier dynamics → surface reactive species → pollutant transformation

This chain is particularly useful for a solid-state physics researcher because it connects measurable physical parameters to chemical function. XRD and microscopy describe the structural state; spectroscopy and electrochemical measurements describe the electronic response; radical and product analysis establish the chemical consequence.

A high photocatalytic rate without evidence of improved carrier separation should not automatically be attributed to a heterojunction effect. Likewise, lower photoluminescence intensity can indicate reduced radiative recombination but does not by itself prove a specific carrier-transfer direction. These distinctions are essential for avoiding overinterpretation.

X. COMPARISON OF MECHANISTIC MODELS:

Criterion	Type-II	Direct Z-scheme	S-scheme	Preferred evidence
Spatial charge separation	Yes	Yes	Yes	PL/TRPL, photocurrent, EIS
Strong redox potentials retained	Often reduced	Yes	Yes	Band energetics + reactive species
Work-function/Fermi-level role	Not defining	Not defining	Central	UPS/KPFM/SPV
Band bending	Not defining	Not defining	Expected	XPS/KPFM
Radical evidence	Supportive	Supportive	Supportive	EPR/scavengers
Single diagram sufficient?	No	No	No	Multiple independent methods

XI. PROPOSED RESEARCH QUESTIONS:

How does controlled defect density alter the band alignment of ZnO/g-C_3N_4?

Can the interface potential be measured directly rather than inferred from photocatalytic activity?

Which characterization combination provides the most defensible distinction between direct Z-scheme and S-scheme interpretations?

How does defect density affect the competition between productive trapping and trap-assisted recombination?

Can photocatalytic activity be normalized using kinetic rate, catalyst mass, optical intensity and mineralization rather than degradation percentage alone?

How stable is the electronic interface after repeated photocatalytic cycles?

XII. EXPECTED OUTCOMES

Because no new experiment has been performed for this manuscript, the following are proposed outcomes rather than reported results:

A measurable relationship between interface quality and charge-transfer behavior is expected.

Controlled defect engineering is expected to alter optical absorption and carrier dynamics, but an optimum defect range rather than unlimited improvement is anticipated.

Combined surface-potential and spectroscopic evidence should provide a more reliable basis for S-scheme claims than band alignment alone.

Mineralization and transformation-product analysis should provide a more meaningful environmental assessment than parent-pollutant removal alone.

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The BICR framework should be transferable to other 2D semiconductor pairs beyond ZnO/g-C_3N_4.

XIII. SUSTAINABILITY AND PRACTICAL CONSIDERATIONS:

A laboratory photocatalyst should not be judged only by its highest degradation percentage. Catalyst synthesis temperature, precursor toxicity, material recovery, long-term stability, light-source energy demand and the fate of transformation products are relevant to practical sustainability. g-C_3N_4 has advantages associated with earth-abundant elements and relatively accessible synthesis, while ZnO is inexpensive and widely available; nevertheless, the complete environmental footprint of a real process depends on synthesis and reactor conditions [5,16].

For future scale-up, reporting should include catalyst loading, pollutant concentration, pH, light spectrum and irradiance, reactor volume and geometry, distance from light source, dissolved oxygen conditions, temperature, sampling protocol and analytical method. Such reporting is necessary for meaningful comparison between laboratories.

XIV. LIMITATIONS OF THE PRESENT WORK

This paper is intentionally presented as a physics-oriented critical perspective and research framework rather than as a claim of new experimental synthesis. It therefore does not report fabricated degradation efficiencies, band gaps, rate constants, photocurrent values or spectroscopy results. The proposed BICR framework must be experimentally validated. In addition, the exact electronic structure of ZnO/g-C_3N_4 depends on synthesis route, crystallinity, defect chemistry, interface orientation and surface termination; consequently, literature values should not be substituted for direct measurements in a future experimental paper.

XV. CONCLUSIONS

ZnO/g-C_3N_4 heterostructures provide a useful model platform for studying the relationship between semiconductor physics and photocatalytic environmental remediation. The central challenge is no longer simply to combine two semiconductors, but to understand and validate how band structure, defects and interface physics govern photogenerated carrier behavior.

A solid-state-physics interpretation requires a chain of evidence connecting band-gap engineering, interfacial charge redistribution, carrier dynamics and surface redox chemistry. Type-II, direct Z-scheme and S-scheme models should therefore be distinguished using complementary experimental methods rather than schematic band diagrams alone. The proposed BICR framework offers a structured route for doing so and provides a natural extension of ZnO/g-C_3N_4 heterojunction research toward mechanism-driven materials design.

The most important future direction is the experimental determination of the interface potential landscape and defect-mediated carrier dynamics, followed by correlation with pollutant transformation and mineralization. Such an approach can help move 2D heterostructured photocatalysts from performance-oriented laboratory studies toward more reproducible, mechanism-validated and sustainable water-remediation technologies.

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