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Interpreting Difference Compton Profiles in Zirconium Hydride: Reference States, Normalization, and Uncertainty

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Abstract
Compton profiles probe projected electron momentum density and can constrain the electronic changes accompanying hydrogen incorporation in zirconium. This focused critical methodological review examines how reference states, normalization, and experimental response affect the interpretation of zirconium-hydride difference profiles. The published Zr/ZrH_2 experiment provides the historical starting point, considered alongside subsequent electronic-structure and spectroscopic evidence. Electron-count conservation distinguishes absolute hydride-minus-metal differences, neutral-hydrogen reference differences, and equal-electron-count shape differences. These constructions answer different questions, and their integrals do not uniquely separate added-electron contributions from host-state rearrangement or determine atom-centered charges. Reproducible analytical examples demonstrate that equal electron counts permit different profile shapes. For an illustrative 40-electron host, metal-reference overscaling of 0.5% reduces the absolute difference integral from 2.0 to 1.8 electrons. Gaussian broadening preserves full-line integrals while attenuating the central feature of the selected redistribution function. An explicit first-order covariance model describes the correlations introduced by uncertain relative normalization. The resulting framework identifies requirements for future comparisons of phase-specific theoretical profiles with composition-controlled measurements, including response matching, finite-window treatment, and joint uncertainty propagation. No historical spectra are refitted, and the analytical examples are neither experimental reconstructions nor first-principles predictions for ZrH_2.

Keywords: Zirconium Hydride (ZrH_2), Protonic Constructions, Host-Electron

Introduction

Hydrogen incorporation changes zirconium in ways that extend beyond the addition of a light interstitial species. The electronic states, equilibrium structure, and local environment evolve together, making zirconium hydride a useful case for examining how a spectroscopic observable supports a statement about bonding. Compton scattering is particularly relevant because it probes electron momentum rather than binding energy or real-space charge directly. A hydride-minus-metal profile can reveal electronic changes that are difficult to isolate in the much larger total scattering signal, but the interpretation of that difference requires an explicit reference state.

Alexandropoulos *et al.* [1] measured difference Compton profiles of Zr and ZrH_2 using 59.54 keV radiation from ^{241}Am and samples of matched optical thickness. They reported agreement with a modified protonic model. That result provides the starting point for the present reassessment. Its continuing significance lies in the sensitivity of a differential momentum-space measurement to hydrogen incorporation, while the interpretation of a successful model comparison should be examined alongside later evidence and the information content of the observable.

The relevant evidence has broadened considerably. Magnuson *et al.* [2] investigated composition-dependent electronic changes in ZrH_x thin films by photoelectron spectroscop$_y$, while Zhang *et al.* [3] considered Zr–H interactions through orbital-based first-principles calculations. In a closely related experimental development, Kothalawala *et al.* [4] measured Ti/TiH_2 Compton profiles in the readily accessible hard X-ray regime. The titanium results renew the methodological relevance of hydride difference profiles, although they cannot establish the corresponding profile or bonding interpretation for zirconium.

This article presents a focused critical methodological review of difference Compton-profile interpretation, using zirconium hydride as its principal case and supporting the discussion with reproducible analytical examples. The interpretive problem is

how to distinguish conclusions fixed by electron-count conservation from those that depend on reference-state selection, experimental processing, and electronic-structure assumptions. The contribution is an integrated framework connecting three commonly used difference constructions, absolute hydride-minus-metal, metal-plus-neutral-hydrogen reference, and equal-electron-count shape differences, to their respective normalization constraints and interpretive limits. The analytical examples demonstrate how profile shape can vary at fixed electron count and how instrumental broadening and relative normalization affect the apparent redistribution signal. These constructions organize the interpretation of electron addition and electronic rearrangement without implying that their separate contributions can be uniquely recovered from a measured difference profile. The framework is used to identify what the published zirconium-hydride evidence supports and what remains unresolved. No new scattering measurements, first-principles momentum-density calculations, or numerical refitting of the 1994 spectra are presented.

Scope and Evidential Basis

The discussion draws on the published Zr/ZrH_2 difference-profile report, established momentum-density theory, zirconium-hydride electronic-structure studies, and recent complementary characterization. It is a focused critical reassessment rather than a systematic review with a predefined search protocol. The literature was selected for its direct relevance to the measured observable, the zirconium–hydrogen electronic system, or experimental constraints on composition and structure. Recent studies are used to update the interpretation rather than to replace the historical evidence with results from a different material.

No channel-level historical counts, complete correction files, or experimental covariance matrices were available for the present calculation. Consequently, the article cannot independently verify the statistical preference for the historical model, determine an updated transfer charge, or infer a revised experimental uncertainty. Statements about that experiment are restricted to its published description and reported conclusion. The numerical examples below are independently defined functions that illustrate normalization and resolution effects; they are neither digitizations nor reconstructions of the published spectrum.

Table 1 identifies the distinct roles of the principal evidence. A measurement of energy-resolved electronic states, an electron-momentum projection, a nuclear-momentum distribution, and a structural measurement constrain different aspects of the same material. Agreement among them becomes informative only when composition, phase, and sample conditions are sufficiently comparable.

Table 1: Published evidence and its role in the reassessment

Study	Primary observable or approach	Relevant constraint
Alexandropoulos *et al.* [1]	Zr/ZrH_2 difference Compton profile	Historical momentum-space comparison and reported model agreement
Magnuson *et al.* [2]	ZrH_x valence-band and core-level photoemission	Composition-dependent hybridization and electronic shifts
Kothalawala *et al.* [4]	Ti/TiH_2 Compton measurement with DFT	Modern comparative test in a related hydride
Zhang *et al.* [3]	First-principles Zr–H orbital analysis	Orbital rearrangement in adsorption mechanisms
Lopez-Honorato *et al.* [5]	Calculated and measured Raman spectra	Hydride characterization and phase assignment
Weick and Grosse [6]	Neutron imaging	Hydrogen content and spatial distribution
Prisk *et al.* [7]	Inelastic neutron scattering	Proton dynamics and nuclear momentum, distinct from electron momentum

Note. Different compositions and sample forms are not assumed equivalent. The present article does not refit any of these datasets.

Information Contained in a Compton Profile

Under suitable high-energy and momentum-transfer conditions, the impulse approximation relates the scattering spectrum to a projection of electron momentum density. The conversion from photon-energy intensity to a Compton profile also requires the appropriate scattering kinematics and experimental corrections. Cooper [8] provides the foundational treatment of these connections and their limits. A measured spectrum should therefore not be identified with an ideal momentum-density projection before the response and correction model has been considered.

In atomic units, let n(p) be the spin-summed electron momentum density, normalized to the electron count of the chosen formula unit. A natural-orbital representation gives:

$$n(p) = \Sigma_\nu f_\nu |\tilde{\varphi}_\nu(p)|^2, \qquad \int n(p)\, d^3p = N \tag{1}$$

Here $\tilde{\varphi}_\nu$ is the momentum-space Fourier transform of a normalized natural orbital, and f_ν is its occupation. In atomic units, the unitary Fourier convention is used: the real-space orbital is integrated against $\exp(-ip\cdot r)$, with the prefactor $(2\pi)^{-3/2}$. Each transformed orbital therefore has a unit integral of its squared magnitude over momentum space, and the sum of the occupations equals N. Equation (1) is exact when the natural orbitals and occupations are those of the exact one-particle reduced density matrix. Replacing them with Kohn–Sham orbitals and occupations gives a practical approximation to the electron momentum density. Reproducing the ground-state real-space density does not, by itself, guarantee an exact momentum distribution.

Taking the scattering-vector direction as z, the ideal electron Compton profile is:

$$J(p_z) = \iint n(p_x, p_y, p_z)\, dp_x\, dp_y, \qquad \int J(p_z)\, dp_z = N \tag{2}$$

All one-dimensional integrals without stated limits in this article extend over the full momentum line. A profile is reported here in electrons per atomic unit of momentum; one atomic unit of momentum is $\hbar/a_0$. For a randomly oriented powder, orientational averaging gives an isotropic profile. In that ideal limit,

$$J(p_z) = 2\pi \int_{|p_z|}^{\infty} n(p)\, p\, dp \tag{3}$$

The radial density can formally be recovered from the derivative of this isotropic profile at nonzero positive momentum. Accordingly, powder averaging does not erase

every feature of the radial momentum distribution. It does erase directional information, and the inversion becomes unstable when derivatives amplify noise or when finite resolution suppresses fine structure. Even an exact radial density does not uniquely specify an atom-centered charge partition, an orbital decomposition, or the real-space wavefunction phases.

The relationship to kinetic energy is also informative. For an isotropic nonrelativistic distribution with adequately represented tails,

$$T = (3/2) \int p_z^2 J(p_z)\, dp_z \quad (4)$$

With momentum expressed in atomic units and the profile normalized to the electron count of the selected formula unit, Equation (4) gives the corresponding nonrelativistic electronic kinetic energy in Hartree atomic units. One Hartree is approximately 27.2114 eV. The factor 3/2 follows from isotropy, which makes the three Cartesian momentum second moments equal. For a randomly oriented powder, the orientationally averaged profile permits this relation even when the underlying crystal momentum density is anisotropic.

Applied to an absolute hydride-minus-metal profile normalized per zirconium atom, the second-moment integral gives the kinetic-energy difference between systems with different electron counts. Dividing that result by the hydrogen content defines a difference per incorporated hydrogen atom but does not isolate the kinetic energy of hydrogen-centered electrons. Changes in the host electronic states and crystal structure also contribute. Moreover, finite momentum coverage, instrumental broadening, and imperfect core cancellation can bias the estimate because the squared-momentum weighting emphasizes the profile tails. Quantitative use therefore requires consistent response treatment and an uncertainty estimate for the unmeasured momentum range.

Reference States and Electron Conservation

The phrase difference Compton profile is incomplete unless its normalization and reference are specified. For neutral material with one zirconium atom per formula unit, Zr contains 40 electrons and ZrH_x contains 40 + x electrons. An absolute difference on this common per-zirconium basis is:

$$\Delta J(p_z) = J_{ZrH_x}(p_z) - J_{Zr}(p_z), \quad \int \Delta J(p_z)\, dp_z = x \quad (5)$$

At x = 2, the full-line integral is two electrons, regardless of whether the electronic rearrangement is described as predominantly ionic, covalent, metallic, or mixed. The area expresses the electron-count difference between neutral systems. It does not identify where the added electronic density resides or how much charge an atom gains under a chosen partitioning scheme.

A different construction subtracts the profiles of neutral hydrogen atoms as well as the metal reference:

$$B(p_z) = J_{ZrH_x}(p_z) - J_{Zr}(p_z) - x\, J_{H\ atom}(p_z), \quad \int B(p_z)\, dp_z = 0 \quad (6)$$

The isolated-atom profile is normalized to one electron. Equation (6) measures departure from a bulk-metal-plus-neutral-hydrogen reference. It contains changes in the zirconium states, the hydrogen-related states, and their hybridization, together with changes caused by the structure adopted by the hydride. It should not be described as a uniquely isolated Zr–H bond contribution. A reference using isolated zirconium atoms would define another quantity.

Equal-area normalization creates a third observable. If valence bookkeeping assigns four electrons to Zr and 4 + x electrons to ZrH_x after a consistent core treatment, an equal-count shape difference is:

$$S(p_z) = [4/(4 + x)]\, J_{val,ZrH_x}(p_z) - J_{val,Zr}(p_z), \int S\, dp_z = 0 \quad (7)$$

The corresponding total-electron expression uses 40/(40 + x). The four-electron valence count reflects the conventional zirconium $4d^25s^2$ bookkeeping; it does not mean that all other electrons are rigorously unaffected by hydrogenation. Semicore treatment must be stated in an actual electronic-structure calculation. Moreover, total-electron rescaling prevents an unchanged core contribution from canceling exactly, because the hydride core is rescaled together with its valence profile.

Table 2 summarizes these distinctions. A zero-area difference need not imply an absence of electronic change. Conversely, a positive area in Equation (5) does not demonstrate that all of the difference can be assigned to hydrogen-centered orbitals. The comparison basis determines which physical question the curve can answer.

Table 2: Reference choices and full-line electron-count constraints

Construction	Reference and normalization	Integral at x = 2
Absolute difference ΔJ	Hydride minus bulk metal; per Zr atom	2 electrons
Neutral-reference difference B	ΔJ minus two neutral H atom profiles	0
Valence shape difference S	Hydride valence rescaled by 4/6; metal unchanged	0
Total-electron shape difference	Hydride total rescaled by 40/42; metal unchanged	0

Note. These identities assume consistent normalization and integration over all momentum. A finite measurement interval does not obey them automatically. A conventional four-electron valence partition is used only where specified.

Real experiments cover finite momentum intervals, so their measured area need not equal the full-line sum rule before extrapolation and correction. Enforcing a target integral over a restricted interval can redistribute a tail error into the central profile. The analytical electron counts in Table 2 should therefore be used as constraints on a complete forward model, with uncertainty in the unmeasured contributions, rather than as automatic normalization targets for an arbitrarily truncated spectrum.

Reassessing Protonic and Bonding Interpretations

A protonic description emphasizes screening of hydrogen nuclei by an electronic system with substantial itinerant character. It should not be read as a claim that a neutral crystal contains unscreened positive charges. Likewise, an anionic description emphasizes electron accumulation around hydrogen, while an atomic reference emphasizes neutral hydrogen-like states. These descriptions become testable only through their specified electronic wavefunctions or predicted observables. Their names alone do not define distinct Compton profiles.

Alexandropoulos *et al.* [1] distinguished three protonic constructions. In “protonic 1,” the added-electron

contribution was proportional to the four-electron zirconium valence profile, retaining its momentum dependence. In "protonic 2," two additional electrons occupied previously empty zirconium band states, raising the band-electron count from four to six within a fixed host potential. This changed the occupied-state momentum distribution rather than merely rescaling the original profile. The calculation used an fcc zirconium reference and reported a Fermi-level increase of 0.10 Ry. "Protonic 3" additionally adopted the hydride lattice constant for the six-electron zirconium calculation. These constructions therefore tested different assumptions about band filling and lattice expansion.

The study compared the experimental difference with atomic, anionic, band-structure, and protonic predictions, broadening theoretical curves with a Gaussian response of 0.59 atomic units FWHM. Its conclusion favored a description based on filling initially unoccupied metal states. This agreement supports the usefulness of that approximation within the reported comparison; it does not uniquely determine hydrogen charge or exclude hybridization. The fcc host approximation and restricted treatment of hydrogen-induced changes must also be considered when interpreting the model's success.

The modern evidence makes a one-component interpretation less satisfactory. Magnuson *et al*. [2] observed a reduction of Zr 4d spectral intensity near the Fermi level with increasing hydrogen content and intensity redistribution toward an H 1s–Zr 4d hybridization region. They also interpreted core-level shifts in terms of charge transfer toward hydrogen. These observations concern specified thin-film compositions and energy-space spectra. They support changes in the zirconium states, but they do not supply a numerical decomposition of a powder Compton difference for bulk stoichiometric ZrH_2.

Hybridization also introduces interference in momentum space. For a schematic normalized orbital $\psi = a\varphi Zr + b\varphi H$, its Fourier intensity contains the cross term $2\,Re[a\,b^*\,\tilde{\varphi}Zr\,\tilde{\varphi}H^*]$ as well as the two diagonal intensities. The resulting profile need not be a simple sum of separately measurable metal and hydrogen atomic profiles. Furthermore, unitary transformations within a subspace of equally occupied orbitals leave the one-particle density matrix, and therefore the total momentum density, unchanged while changing the individual orbital contributions. This invariance does not generally apply to arbitrary mixing of orbitals with unequal occupations if their occupation weights are held fixed. An orbital decomposition is therefore representation dependent, whereas the total Compton profile is determined by the underlying one-particle density matrix. Chemical assignments based on selected orbitals or real-space charge partitions should consequently be distinguished from quantities directly constrained by the measured profile.

Zhang *et al*. [3] examine Zr–H interactions through d-orbital rearrangements in a first-principles treatment of adsorption mechanisms. Their work adds a recent orbital perspective, but an adsorption calculation should not be treated as a measured bulk Compton profile. The implication for reassessment is methodological: candidate models should allow the host states to reorganize instead of assuming that hydrogen merely contributes an unchanged atomic profile to an unchanged metal.

Kothalawala *et al*. [4] provide a contemporary comparison through measurements of Ti and TiH_2 using two detector systems and density-functional calculations. Their approximately parabolic difference profile supports an itinerant screening interpretation. However, the experimental profiles were normalized over a finite momentum interval using the corresponding theoretical integrals, and cancellation of multiple-scattering contributions was supported by assumptions about sample similarity and the broad character of those contributions.

For the present methodological framework, these details mean that the reported agreement should be interpreted together with the normalization and correction assumptions. This observation does not invalidate the titanium result; it identifies conditions to examine when applying a similar analysis to zirconium hydride. A renewed Zr/ZrH_x comparison should test whether candidate models remain distinguishable when uncertainties in normalization, response, composition, and residual corrections are included. The titanium evidence therefore supports the continuing relevance of difference-profile measurements without independently establishing the corresponding zirconium bonding description.

Analytical Demonstration of Profile Ambiguity

A reproducible example makes the reference-state distinctions explicit. The one-electron profile of an isotropic ideal Fermi sphere with radius p_F is:

$$F(p_z; p_F) = [3/(4p_F)]\,[1 - (p_z/p_F)^2] \text{ for } |p_z| \le p_F;\ 0 \text{ otherwise} \quad (8)$$

Its integral is one. A neutral hydrogen atom in the nonrelativistic 1s ground state has the normalized profile:

$$J_{H\ atom}(p_z) = [8/(3\pi)]\,(1 + p_z^2)^{-3} \quad (9)$$

These functions are analytical reference shapes. Neither is introduced as a complete band-structure model of zirconium hydride.

The illustrative metal valence profile is $4F(p_z; 1.0)$. The constructed hydride-minus-metal difference is:

$$\Delta J_{demo}(p_z) = 2F(p_z; 1.2) + R(p_z) \quad (10)$$

$$R(p_z) = 0.4\,[G(p_z; 1.1) - G(p_z; 0.45)] \quad (11)$$

Here G is the normalized, zero-mean Gaussian defined by:

$$G(p_z;\ \sigma) = \frac{1}{\sqrt{2\pi}\sigma}\ exp(-\frac{p_z^2}{2\sigma^2})$$

Where σ is the standard deviation in atomic units of momentum. Thus, the values 1.1 and 0.45 in Equation (11) denote standard deviations, whereas the instrumental response widths discussed below are specified as full widths at half maximum. The Gaussian has a unit full-line integral. Consequently, the first term in Equation (10) supplies two electrons, while R transfers profile weight from lower to higher momentum without changing the integrated electron count. The coefficient 0.4 is an illustrative weight expressed in electrons; it is neither a fitted charge-transfer value nor a measured hydrogen population.

This separation is a chosen analytical construction rather than a uniquely identifiable physical decomposition. Any alternative addition function with an integral of two electrons could be subtracted from the same absolute

difference to define another zero-area residual. Electron-count conservation therefore fixes the total difference integral but does not uniquely distinguish an added-electron profile from rearrangement of the host states. Assigning these contributions requires additional electronic-structure assumptions or independently constrained calculations. In particular, R should not be interpreted as an experimentally isolated host-electron contribution.

The constructed total hydride profile remains nonnegative everywhere, satisfying a necessary consistency condition for a Compton profile. This check does not establish that the assumed functions represent the electronic structure of ZrH_2. Fig 1a compares three functions: twice the isolated-hydrogen 1s profile, twice the normalized ideal Fermi-sphere profile, and the constructed addition-plus-redistribution difference. Each has a full-line integral of two electrons, although their central amplitudes and tails differ. Equal electron counts therefore do not determine profile shape. Fig 1b compares the redistribution term R, the neutral-reference difference B, and the valence equal-count shape difference S. Each has zero full-line integral, but they represent different constructions and their positive and negative regions have different interpretations. All curves are analytical illustrations rather than measured or first-principles ZrH_2 profiles. The displayed momentum interval is narrower than the integration interval used for the numerical checks.

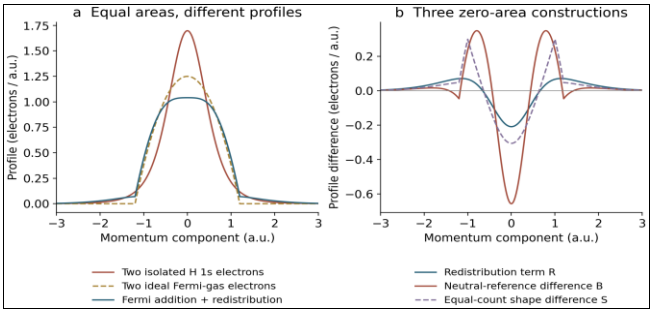


Fig 1: Analytical profile comparisons: (a) three functions with full-line integrals of two electrons; (b) the zero-area constructions R, B, and S. Definitions and interpretive limits are given in the text

The central value of the constructed absolute difference is 1.04045 electrons per atomic unit, whereas two isolated hydrogen 1s profiles give 1.69765. This contrast is a consequence of the chosen functions. It does not estimate the experimental difference between hydrogen states in ZrH_2 and atomic hydrogen. The numerical example establishes only that equal electron counts do not determine profile shape and that a zero-integral redistribution can modify an addition profile appreciably.

The redistribution function R provides a controlled example of instrumental broadening. Convolution with a unit-area Gaussian response preserves its zero full-line integral while changing its local amplitudes and shape. For the selected parameters, response full widths at half maximum of 0.4 and 0.8 atomic units reduce the magnitude at zero momentum from 0.20955 to 0.18840 and 0.14441 electrons per atomic unit, respectively, corresponding to reductions of approximately 10.1% and 31.1%. These percentages describe this particular function and are not general attenuation factors for zirconium-hydride redistribution.

The response widths were selected to illustrate sensitivity to broadening; they were neither fitted to experimental data nor used to reconstruct the historical instrument response. Likewise, the Gaussian widths and redistribution coefficient define a mathematical example rather than a calibrated electronic model. The calculation therefore establishes that broadening can weaken a central feature without changing the full-line electron-count constraint, but it does not quantify the suppression of any feature in the published Zr/ZrH_2 experiment.

Area preservation also requires integration over the full momentum line. The figures display only a restricted interval, whereas the numerical checks use the wider interval specified in the appendix. Broadening can move profile weight across the boundaries of a finite interval, so an integral evaluated only over the plotted or measured window need not remain unchanged. Any experimental application must therefore account for the unmeasured tails and their uncertainties.

Experimental Response and Uncertainty

A quantitative comparison should begin with a forward model linking each intrinsic Compton profile to the corresponding measured quantity. For each specimen, this model should include the intensity scale, instrumental response, background, and corrections required to convert the scattering spectrum into a momentum profile. A convolution description is appropriate only when the response can reasonably be treated as shift invariant over the momentum range considered. Otherwise, a response matrix or an energy-domain measurement model is required. Matching the optical thickness of the metal and hydride specimens helps control some experimental contributions but does not establish that their responses and correction uncertainties are identical.

For profiles placed on a common absolute scale, a shared linear convolution operator commutes with subtraction. The measured difference is then the intrinsic hydride-minus-metal difference broadened by that common response. When the responses differ, the subtraction also contains a contribution from their mismatch acting on the underlying profiles. Because those profiles contain the much larger host-electron contribution, even a small mismatch can generate a feature comparable to the desired difference signal. An unchanged intrinsic core therefore need not cancel in the measured subtraction.

Candidate metal and hydride profiles should consequently be propagated separately through their respective measurement models before subtraction. Uncertainty in response width, momentum calibration, background, and relative scale should be propagated jointly with the profile uncertainties. This procedure allows a proposed redistribution feature to be assessed against plausible experimental distortions without assuming that subtraction removes them completely.

Relative normalization is especially important because the desired difference is small compared with the host profile. If the metal reference is overscaled by a fractional amount ε,

$$\Delta J_{\varepsilon}(p_z) = J_{ZrH_2}(p_z) - (1 + \varepsilon)J_{Zr}(p_z) = \Delta J(p_z) - \varepsilon J_{Zr}(p_z) \quad (12)$$

For full total-electron profiles, the difference integral becomes $2 - 40\varepsilon$. A 0.1% metal overscaling therefore changes this integral by 0.04 electron, or 2% of the two-

electron addition. This is an algebraic sensitivity statement, not an estimate of the normalization uncertainty in any published experiment.

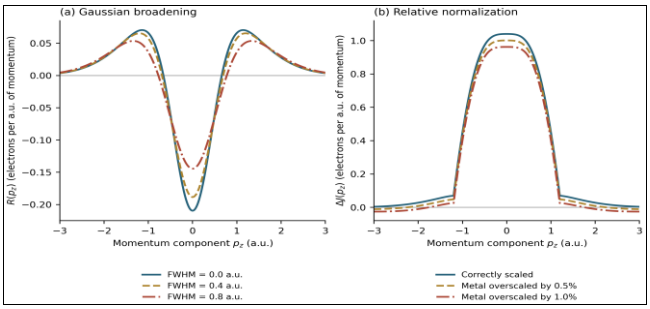


Fig 2: Analytical sensitivity examples: (a) Gaussian broadening of the redistribution term R; (b) distortion of the absolute difference caused by metal-reference overscaling. Response widths and scale deviations are identified in the legends

For Fig 2, the analytical metal profile includes an assumed 36-electron Gaussian core with a standard deviation of 3.0 atomic units of momentum, in addition to the four-electron valence term. The same core is included in the constructed hydride, so it cancels under correct subtraction. Overscaling the metal by 0.5% or 1.0% changes the full-line difference area to 1.8 or 1.6 electrons. The respective central errors are −0.03894 and −0.07787 electrons per atomic unit.

Table 3 summarizes the broadening and normalization examples in two groups. For Gaussian broadening, the central values refer to the redistribution function R at zero momentum, while its full-line integral remains zero. For metal-reference overscaling, the central values instead represent errors relative to the correctly scaled absolute difference, rather than the resulting profile amplitudes. The corresponding full-line integrals decrease from two electrons as the metal-reference scale increases. The assumed Gaussian core is a bookkeeping device and does not reproduce zirconium atomic-core momentum structure.

Fig 2 illustrates two distinct effects. Panel a shows how Gaussian broadening changes the amplitude and shape of the zero-area redistribution term R. Panel b shows how overscaling the metal reference alters the absolute hydride-minus-metal difference. In the latter construction, both specimens contain the same assumed Gaussian core contribution, which cancels under correct subtraction but leaves a residual when the metal is overscaled. The resulting distortion therefore includes a core contribution as well as a valence contribution. These examples illustrate measurement sensitivities; they do not estimate errors in the historical experiment.

Table 3: Effects of Gaussian broadening and metal-reference overscaling in the analytical example

Scenario	Central value or error (electrons/a.u.)	Full-line area (electrons)
Gaussian FWHM 0.0 a.u.	-0.20955	0
Gaussian FWHM 0.4 a.u.	-0.18840	0
Gaussian FWHM 0.8 a.u.	-0.14441	0
Metal overscaling 0.1%	-0.00779	1.96
Metal overscaling 0.5%	-0.03894	1.80
Metal overscaling 1.0%	-0.07787	1.60

Note. All profile parameters, response widths, and scale deviations are illustrative assumptions, not estimates derived from experimental data. Integrals refer to the full momentum line.

Normalization uncertainty introduces correlations between momentum bins because the same uncertain reference scale affects the entire difference profile. Let the measured hydride and metal profile vectors be H and M, with covariance matrices C_H and C_M, and define their difference as:

$$D = H - aM$$

Where a is the relative metal-reference scale. If a is statistically independent of both profile vectors, first-order uncertainty propagation gives:

$$C_D \simeq C_H + a^2 C_M - a(C_{HM} + C_{HM}^T) + u^2(a) m m^T$$

Here C_{HM} = Cov(H,M), u(a) is the standard uncertainty of the scale, and m is the expected metal-profile vector, estimated in practice by the reference profile. The scale and profile values on the right-hand side are evaluated at their nominal estimates. The final term is an outer product: its element for bins i and j is $u^2(a) m_i m_j$. Consequently, scale uncertainty produces a coherent distortion across the profile rather than independent fluctuations in individual bins. Shared calibration, background, or correction inputs may also make C_{HM} nonzero, even when the counting statistics of the two acquisitions are independent.

This expression is a first-order approximation. If the scale is estimated by normalizing or fitting the same profiles, its independence cannot be assumed. The additional correlations must then be included by propagating the complete measurement and normalization procedure through its Jacobian or through joint Monte Carlo sampling. Treating a fitted scale as an independent uncertainty input can misrepresent the uncertainty of the resulting difference.

The uncertainty framework of the Joint Committee for Guides in Metrology [9] supports propagating these inputs through an explicit measurement model. In a future fit, model residuals should be evaluated with the covariance retained on an independent data subspace. Exact area normalization can make a covariance matrix singular, while smoothing or interpolation can reduce the effective number of independent observations. A visually satisfactory residual curve is therefore insufficient to establish a statistically meaningful preference for one bonding model.

Structural and Computational Constraints

An electronic reassessment must specify which zirconium hydride is being compared with which metal reference. The designation ZrH_x covers different compositions, structures, and possible mixtures. Quijano *et al.* [10] explicitly examined the electronic structure and energetics of tetragonal distortion in transition-metal dihydrides including ZrH_2. Their work reinforces the need to evaluate the relevant structure rather than assume that a convenient cubic cell represents every hydride condition.

A useful computational comparison would separate hydrogen incorporation from changes in the host geometry. It could compare metal at its own equilibrium structure, zirconium constrained to a hydride-like lattice, and the complete hydride at the same lattice. Such a decomposition is a controlled theoretical path, not a unique chemical partition; the intermediate metal structure may be metastable or mechanically unstable. Its value is to reveal whether a

predicted momentum change arises primarily from geometry, hydrogen-related hybridization, or their coupling.

The zirconium-specific calculations of Papanicolaou *et al.* [11] provide a direct theoretical foundation for interpreting hydrogen-induced momentum changes. Using a self-consistent augmented-plane-wave method, they calculated electron momentum distributions in Zr and ZrH_2 and examined their behavior along the [100], [110], and [111] directions. They related the changes accompanying hydrogen incorporation to the corresponding energy bands and Fermi-surface topology. Their work therefore supplies a material-specific account of momentum redistribution that goes beyond adding an isolated-hydrogen profile or rescaling the metal profile.

For the present interpretation, these calculations and the simplified protonic constructions should be treated as different levels of electronic description. A useful, renewed comparison would determine which predicted differences survive powder averaging and experimental broadening, and whether those surviving features distinguish changes in band occupation from the fuller electronic reorganization accompanying hydride formation. The present analytical examples do not perform that comparison.

A modern calculation must evaluate electron momentum densities and their projections explicitly. A density of states groups states by energy and cannot alone determine a Compton profile. Ernsting *et al.* [12] describe a practical approach using tetrahedron interpolation. Application to ZrH_x would require phase-specific structures, converged wavevector sampling, momentum cutoffs, occupations, and orientation averaging, together with consistent core and semicore treatment.

The distinction between practical Kohn–Sham momentum densities and the interacting momentum distribution should also be retained. Correlation corrections, if introduced, must conserve the relevant electron count and be applied consistently to metal and hydride. An unconverged high-momentum tail or inconsistent reconstruction of all-electron wavefunctions can create apparent redistribution. Agreement of total energies or lattice parameters alone does not verify the corresponding momentum profile.

Temperature and phase conditions also matter. Danakas *et al.* [13], in an experimental and theoretical Compton study of calcium at room and high temperature, provide a relevant example of comparing electronic momentum profiles under different material conditions. This does not establish the magnitude of a thermal effect in zirconium hydride. It does underscore the need to match the physical state of the measurement and calculation before a residual is assigned exclusively to hydrogen bonding.

Complementary Measurements and Future Reassessment

Composition and structure should constrain the profile interpretation independently wherever possible. Lopez-Honorato *et al.* [5] combined calculated Raman spectra with measurements to characterize zirconium hydride and identify hydride-related signals. Raman characterization can therefore help constrain which phases a Compton model should represent, while diffraction remains necessary for lattice parameters, texture, and phase fractions. Neither measurement directly supplies the electronic momentum redistribution.

Weick and Grosse [6] discuss neutron imaging as a method for investigating hydrogen content and distribution in zirconium alloys. Spatial hydrogen information is valuable because a bulk-averaged Compton profile can combine metal-rich and hydride-rich regions. For a multiphase specimen, theoretical profiles must be weighted on a declared basis, such as fractions of zirconium atoms in each phase. A volume fraction cannot be inserted as an electron-count fraction without conversion using the appropriate densities and compositions.

Electron and neutron Compton profiles must also be distinguished. Prisk *et al.* [7] measured proton dynamics and momentum distributions in zirconium-containing hydrogen systems. Those nuclear momentum distributions concern hydrogen motion and its effective potential, rather than the electron momentum density in Equation (2). They provide complementary information about hydrogen dynamics but cannot substitute for the electronic profile or determine electron transfer by direct integration.

A substantive experimental reassessment would begin with independently characterized ZrH_x specimens, suitable metal references, and an instrument response measured under the same acquisition conditions. The analysis should retain channel counts, sample geometry, normalization information, backgrounds, and correction uncertainties. Candidate theoretical profiles should then be propagated through the same forward model. Their distinguishability should be tested after allowing justified uncertainty in composition, response, and scale, rather than using those nuisance parameters to absorb any disagreement.

Directional measurements on sufficiently characterized material could add information lost in a powder average. Where such measurements are impractical, several compositions with independently determined phase content could still constrain the evolution of a common theoretical description. Cross-validation against photoelectron spectra or related observables should compare calculated observables directly, since an apparent agreement between two qualitative bonding labels does not establish quantitative consistency.

The result of such a program could be either confirmation or revision of the earlier interpretation. The present reassessment does not predetermine that outcome. It identifies the conditions under which a modified screening description, a hybridized band description, or a more elaborate treatment becomes distinguishable through momentum-space evidence.

Conclusions

A zirconium-hydride difference Compton profile contains the effects of added electrons and the redistribution of the electronic system already present in the metal. Its physical interpretation depends on the reference state. The absolute per-zirconium difference integrates to x electrons for neutral ZrH_x, while a metal-plus-neutral-hydrogen reference difference and an equal-count shape difference each have zero area. These identities distinguish observables; they do not assign atom-centered charges.

The published historical agreement with a modified protonic model remains an important result, but it should be interpreted alongside subsequent evidence for Zr–H hybridization and host-state rearrangement. Itinerant screening and local charge redistribution can coexist. A

powder profile constrains their combined radial momentum signature without uniquely resolving their orbital or atomic partition.

The methodological contribution of this review is to connect reference-state definitions, electron-count constraints, and experimental uncertainty within a common framework for interpreting difference Compton profiles. The analytical examples demonstrate that equal integrated counts do not determine profile shape, that broadening can suppress a redistribution feature while preserving its full-line integral, and that relative normalization errors introduce correlated distortions. These results identify interpretive requirements rather than establish a revised electronic structure for zirconium hydride. A future quantitative reassessment should compare phase-specific theoretical profiles with composition-controlled measurements through consistent forward models, retaining uncertainty in response, normalization, corrections, and unmeasured tails. Whether such a comparison confirms or modifies the historical protonic interpretation remains an open question.

Data and Code Availability

No new experimental data were collected, and no published spectrum was digitized or refitted. Supplementary Material S1 provides the Python script *Supplementary_S1_Compton_Profiles.py*, which generates both analytical figures and reproduces the numerical examples summarized in Table 3. The script includes electron-count checks, response convolution, reference-scale sensitivity calculations, plotting instructions, and software-version reporting. Its output file, *results.json*, records the parameters and numerical results. These calculations illustrate mathematical properties of profile differences and are not validated electronic-structure predictions for ZrH_2.

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