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Supplementary Information for: Domain-specific ChatBots for Science using Embeddings

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1 Examples of Chatbot Performance

Below we provide examples of chatbot responses to various scientific questions. We compare unaided responses (i.e. without providing any text chunks from the document store) to contextualized prompts (i.e. where text chunks are added to the prompt). We also compare the GPT-3.5 model (accessed via the OpenAI API) and ChatGPT-4 (accessed via the web interface). The LLM responses are unedited, other than reformatting to convert symbols into IATEX. Highlights were added to the response text. Green highlights show correct responses highly-matched to the user query. Red text highlights erroneous parts of the response (highly mismatched to user needs or delivering incorrect information). Reasonable and factually correct (though not always optimal or necessary) parts of the response are left unhighlighted.

1.1 Example: Answer scientific question

User Query: How can I use x-ray scattering to measure the grain size?

• GPT-3.5 (unaided) response:

X-ray scattering can be used to measure the grain size of a material by analyzing the diffraction pattern produced when a beam of X-rays interacts with the material's crystalline lattice structure. As the X-rays pass through the material, they are diffracted by the crystal planes and produce a pattern of peaks that can be analyzed to determine the crystal structure and grain size.

According to a paper published in Materials Science and Engineering: R: Reports, " The grain size can be calculated using the Scherrer equation, which relates the peak width to the grain size, or by more advanced methods such as the Warren-Averbach method or the Williamson-Hall method." [1]

There are also several specialized X-ray diffraction techniques, such as small-angle X-ray scattering (SAXS) and grazing-incidence small-angle X-ray scattering (GISAXS), that can provide additional information on the size and shape of nanocrystals and thin films.

Reference: [1] Salgado-Morán, G. et al. (2017). In situ and Operando X-ray Techniques in Nanomedicine: From Preclinical Development to Advanced Clinical Applications. Materials Science and Engineering: R: Reports, 114, 1-46. https://doi.org/10.1016/j.mser.2016.10.001

Analysis: Fabricated ("hallucinated") a non-existent reference. Did not directly explain the method/calculation. **Poor response**.

• GPT-3.5 (with context chunks) response:

X-ray scattering is a powerful technique that can be used to measure structural order at the atomic/molecular scale through wide-angle X-ray scattering (WAXS) and at the nanoscale through small-angle X-ray scattering (SAXS). X-ray scattering can be used to measure the average structure throughout a material, making it complementary to real-space imaging techniques, which may provide only local measurements difficult to generalize.

To measure grain size using X-ray scattering, one can use conventional X-ray powder diffraction measurements, which yield distinct rings on the detector. The ring position, $q = (4\pi/\lambda)\sin\theta$, provides information about the spacing of structures $(d = 2\pi/q)$. The average grain size can be estimated from the peak width. One method is the Debye-Scherrer analysis, which allows extraction of a coherence length from the peak width using $\lambda/\Delta\lambda \approx 2K/q$, where the Scherrer constant K is of order unity.

There is a peak width limit for the Scherrer analysis, however, meaning that the grain size that can be measured with this method is typically limited to the submicron range for modern synchrotron beamlines. An alternative method called "variance scattering" has been proposed, which quantifies the statistical variation of intensity along a scattering ring. This method enables quantification of grain size across a wide range of sizes, from nanometers to hundreds of microns, making it useful for materials with extremely large or micrometer-sized grains.

Analysis: Explained method using a partially incorrect equation. Provided information about alternate methods from source documents. **Partial success**.

• ChatGPT-4 (unaided) response:

X-ray scattering techniques, such as X-ray diffraction (XRD), can be used to measure the grain size in a polycrystalline material. This is often achieved through the application of the Scherrer Equation, which relates the size of the crystallites in a sample to the broadening of the diffraction peak.

The key steps in this process include:

1. **X-Ray Diffraction Experiment**: The first step is to conduct an XRD experiment on your sample. The sample is bombarded with X-rays and the diffraction pattern is recorded.

2. **Peak Identification**: Identify the diffraction peaks in the XRD pattern. These peaks correspond to different planes of atoms in the crystalline material, and the peak broadening is related to the grain size.

3. Measure Peak Broadening: The width of the diffraction peak at half its maximum height, known as the Full Width at Half Maximum (FWHM), is measured. This is typically done in radians.

4. Application of Scherrer Equation: The Scherrer Equation is typically written as:

$\tau = K\lambda/(\beta \cos\theta)$

Where:

- $-\tau$ is the crystallite size
- $-\ K$ is the shape factor, with a usual value of about 0.9, but it depends on the specific crystallite geometry
- λ is the X-ray wavelength
- $-~\beta$ is the line broadening at half the maximum intensity (FWHM), after subtracting the instrumental line broadening, in radians
- $-\theta$ is the Bragg angle (the angle between the incident ray and the scattering planes)

Note that the Scherrer Equation assumes that the peak broadening is only due to the crystallite size, but in reality, there could be other factors contributing to the broadening, such as strain within the material. So, the grain size calculated using this method should be taken as an approximation. More accurate methods would involve additional corrections or use more advanced techniques such as electron backscatter diffraction (EBSD).

Also remember that this method provides the average size of crystallites that are contributing to the diffraction peak, not necessarily the average grain size of the entire sample, as not all grains contribute equally to the XRD pattern.

Analysis: Detailed and accurate answer, using general knowledge. Provided additional information and caveats. **Good response**.

• ChatGPT-4 (with context chunks) response:

X-ray scattering is a powerful technique to measure grain size in various materials. The fundamental concept is that when X-rays are incident on a crystalline material, they are diffracted in different directions based on the periodic arrangement of atoms. This diffraction pattern contains information about the size of the grains within the material.

There are two primary methods of grain size estimation using X-ray scattering mentioned in the sources you provided:

1. Scherrer analysis: This method uses the peak width of the diffracted X-ray pattern. The broadening of these peaks provides information about the average size of the crystalline domains or 'grains' in the material. The Debye-Scherrer equation $(D = K\lambda/\beta\cos\theta)$ can be used, where D is the crystallite size, K is the shape factor (Scherrer constant), λ is the X-ray wavelength, β is the full width at half maximum (FWHM) of the peak, and θ is the Bragg angle. However, this method has limitations and can usually only measure submicron grain sizes, as instrumental effects such as beam divergence impose a minimum peak width.

2. Graininess or non-uniformity method: This newer method, discussed by Yager, Majewski, et al., relies on quantifying the non-uniformity of X-ray scattering rings, which arise from the finite number of grains in the probed volume. In essence, if you have many small, randomly-oriented grains, they give rise to a uniform scattering ring as the numerous Bragg peaks overlap. In contrast, if you have a smaller number of larger grains, you get a scattering ring with significant intensity variation. By looking at this variance or non-uniformity in the scattering ring, you can estimate the number of grains and thus the grain size, over a wide range from nanometers to hundreds of microns. Importantly, this method can be used even when the grains are too large for a conventional Scherrer analysis, when the intrinsic peak width is much smaller than the instrumental broadening.

So, depending on the size of the grains you are interested in and the specifics of your sample and experimental setup, you can choose either the Scherrer analysis or this new method based on non-uniformity of scattering rings to measure grain size using X-ray scattering.

Reference:

- Yager, Majewski, et al. "Metrics of graininess: robust quantification of grain count from the non-uniformity of scattering rings"

- Majewski, Yager, et al. "Rapid ordering of block copolymer thin films"

 $\label{eq:Analysis: Correctly synthesized provided information, comparing and integrating with established information. Excellent response.$

1.2 Example: Describe specific concept

User Query: Describe the core novelty of GTSAXS

• GPT-3.5 (unaided) response:

GTSAXS stands for Grazing-incidence through-Silicon-etched Anisotropic X-ray Scattering and it is a nondestructive analytical technique that can be used to study the microstructure of thin films and interfaces.

According to a paper published in the Journal of Applied Crystallography by Dima et al. (2021), the core novelty of GTSAXS lies in its ability to probe in-plane microstructures by exploiting the anisotropic scattering from etched surfaces in silicon. In other words, by measuring the scattering of X-rays at a grazing incidence angle, the technique provides information about the orientation and arrangement of the microstructure within the plane of the film. This is particularly useful for studying the properties of anisotropic materials such as liquid crystals or aligned carbon nanotubes.

Overall, GTSAXS represents a valuable addition to the analytical toolbox for studying thin films and interfaces, providing complementary information to other techniques such as X-ray reflectivity or transmission electron microscopy.

Analysis: Fabricated citation and associated results. Bad response.

• GPT-3.5 (with context chunks) response:

Grazing-incidence transmission small-angle X-ray scattering (GTSAXS) is a new method for determining the three-dimensional order in nanostructured thin films, and its core novelty lies in its ability to eliminate the multiple scattering and refraction issues associated with typical grazing-incidence analysis, while still retaining the sensitivity of grazing-incidence small-angle X-ray scattering (GISAXS) to lateral and perpendicular film structure. GTSAXS requires a focused beam, which illuminates a region near the edge of the sample, so that the scattering can exit from the edge of the substrate, and this arrangement substantially reduces the multiple scattering events that complicate GISAXS data analysis, especially at small surface-normal scattering vectors. Importantly, GTSAXS is capable of probing very small and negative surface-normal scattering vectors, which is particularly well-suited for investigating structural features of anisotropic materials with a film thickness comparable to the inverse-surface-normal scattering vector magnitude.

Analysis: Correctly identified relevant paper, and summarized it. Good response.

• ChatGPT-4 (unaided) response:

As of my last training cut-off in September 2021, the term "GTSAXS" doesn't refer to any widely known technique, concept, or innovation in any field I'm familiar with, including technology, computer science, physics, chemistry, biology, or any other discipline. If "GTSAXS" refers to a more recent development or an abbreviation that isn't commonly used or recognized, I may not have the information on it.

Generally, SAXS in scientific contexts often refers to Small-Angle X-ray Scattering, a technique used to determine the structure of materials at a nanoscale level. However, the "GT" prefix isn't standard and might stand for a specific adaptation or application of the technique, but I don't have the specific information about that.

I recommend checking the most recent resources or directly asking the organization or researchers that use this term.

Analysis: Acknowledged lack of information. Made reasonable inferences. Valid response.

• ChatGPT-4 (with context chunks) response:

Grazing-incidence transmission small-angle X-ray scattering (GTSAXS) is a novel method for analyzing the three-dimensional order in nanostructured thin films. This method represents a unique blending of the advantages of GISAXS (Grazing-incidence small-angle X-ray scattering) and TSAXS (Transmission small-angle X-ray scattering). It has the capacity to probe very small and even negative surface-normal scattering vectors, making it particularly useful for studying structural features when the film thickness is comparable to the inverse-surface-normal scattering vector magnitude.

There are several key aspects of GTSAXS that make it innovative:

1. GTSAXS retains the sensitivity of GISAXS to lateral and perpendicular film structure while eliminating multiple scattering and refraction issues typically associated with grazing-incidence analysis. This is achieved by the unique arrangement of a sample/beam geometry where the scattering exits through the edge of the substrate, resulting in scattering images that align with straightforward (Born approximation) scattering models. 2. The GTSAXS geometry pushes distortion away from the region of interest, resulting in only a small and slowly varying correction, which can be made arbitrarily small by choosing a sufficiently large incident angle. The correction is dependent only on the known substrate density, making it less sensitive to the film density.

-3. The technique requires a focused beam that illuminates a region near the edge of the sample, as the scattering must exit from the edge of the substrate. This substantially reduces the multiple scattering events that complicate GISAXS data analysis.

4. Another significant advantage of GTSAXS is the straightforward analysis within the Born approximation framework. By combining GTSAXS data at small q z and GISAXS data at larger q z, where refraction effects are not severe, it's possible to obtain surface scattering results that incorporate the best aspects of both techniques.

The method was presented and discussed in the paper "Grazing-incidence transmission X-ray scattering: surface scattering in the Born approximation" by Lu, Ocko, et al.

Analysis: Detailed response, drawing out the most significant results from the paper. Provides valid citation. **Excellent response.**

1.3 Example: Describe specific result, based on vague description

User Query: What new morphologies were found for BCP blend thin films?

• GPT-3.5 (unaided) response:

There have been several studies that have explored new morphologies of block copolymer (BCP) blend thin films. One recent example is a study published in the journal Polymer, where researchers investigated the use of solvent vapor annealing to induce morphological transitions in thin films of a blended BCP composed of poly(styrene-b-methyl methacrylate) (PS-b-PMMA) and polystyrene-block-poly(vinylpyridine) (PS-b-P4VP). The authors showed that by exposing the thin films to different solvents, they were able to induce the formation of a range of different morphologies, including lamellar, cylinder, sphere, and gyroid structures. Another study published in the journal ACS Applied Materials & Interfaces discussed the use of graphene oxide as a substrate for BCP thin films. The authors found that by varying the thickness of the graphene oxide substrate, they were able to induce the formation of a range of different morphologies in the BCP films, including cylinders, spheres, and gyroids.

Analysis: Information is not properly sourced, and may be fabricated and/or not related to blends. Bad response.

• GPT-3.5 (with context chunks) response:

The provided snippets do not include information on new morphologies found for BCP blend thin films. However, it is mentioned that blending BCPs with other species, including other BCPs, can dramatically alter the morphology that forms during self-assembly, and that the behavior of BCP/BCP blends in thin films remains largely unexplored. [From Bae, Yager, et al. "Chain Redistribution Stabilizes Coexistence Phases in Block Copolymer Blends"]

Analysis: Did not provide concrete examples of new morphologies. Poor response.

• ChatGPT-4 (unaided) response:

I'm sorry for the inconvenience, but as of my knowledge cutoff in September 2021, I don't have any updated information on the specific topic of new morphologies for Block Copolymer (BCP) blend thin films after that date. Block Copolymers (BCPs) can form a variety of morphologies in thin films, such as lamellae, cylinders, spheres, gyroids, etc., due to the microphase separation between the incompatible polymer blocks.

To get the most accurate and updated information, I recommend referring to the latest scientific literature in the field, such as academic articles, research papers, or reviews on the subject. These could be found in scientific databases like PubMed, IEEE Xplore, ScienceDirect, or the American Chemical Society's publications.

If you need help in understanding the principles behind BCP blend thin films, I would be glad to help with that based on the information available up to 2021.

Analysis: Acknowledged lack of information. Valid response.

• ChatGPT-4 (with context chunks) response:

Recent studies on Block Copolymer (BCP) blend thin films have revealed different morphologies, primarily influenced by factors like BCP concentration, film thickness, substrate topography, and annealing processes. The studies reported new observations on how morphologies change due to various factors, rather than entirely new morphologies per se.

In the study by Basutkar, Yager, et al., the BCP thin films usually adopted a horizontal cylinder morphology in the continuous film region. However, near the edges of the film, the morphology becomes anti-aligned. Interestingly, in regions scribed to yield multiple edges near one another, a hexagonal packing morphology (vertical cylinders) was observed due to multiple competing directing forces ("Aligned Morphologies in Near-Edge Regions of Block Copolymer Thin Films").

In Rahman, Yager, et al.'s work, it was found that film thickness and substrate topography can control the registration of a BCP film, leading to a variety of 3D morphologies of BCP materials ("Non-native three-dimensional block copolymer morphologies"). They discussed the "responsive layering" of block copolymer nanostructures, which can generate a wide variety of 3D morphologies. In some cases, vertical orientation of morphology was observed in monolayer or sub-monolayer films.

Zhang, Karim, et al. also discussed the strong effect of film confinement on the BCP morphology, indicating that the surface morphology of these model block copolymer films is influenced by film thickness and annealing temperature ("Surface Morphology Diagram for Cylinder-Forming Block Copolymer Thin Films"). In another study, Zhang, Douglas, et al. observed a thermally reversible surface order–order transition, confirming that this transition is independent of film thickness in the range of 40–170 nm ("Thermally Reversible Surface Morphology Transition in Thin Diblock Copolymer Films").

In general, these studies provide insight into the morphologies that BCP blend thin films can adopt under different conditions and contribute to a better understanding of the behavior and potential applications of these materials.

Analysis: Incorrectly assumed that provided text chunks were all referring to blend studies. Provided summaries are correct, but not related to the user question. **Bad response**.

1.4 Example: Describe specific results

User Query: Were novel morphologies found when ordering blends of block copolymers (BCP) chains using directed self-assembly (DSA); i.e. by ordering using chemical strip patterns? If so, what new morphologies were discovered?

• GPT-3.5 (unaided) response:

Yes, novel morphologies have been discovered when ordering blends of block copolymers (BCP) chains using directed self-assembly (DSA) with chemical strip patterns.

Research has shown that specific patterns of chemical strips can be used to direct the self-assembly of block copolymer chains, resulting in the creation of new, complex morphologies. For instance, a group of researchers used a combination of chemoepitaxy and graphoepitaxy to demonstrate the fabrication of a new morphology, which they called "bottlebrush-on-tape." This morphology was formed by the directed assembly of a polystyrene-b-polydimethylsiloxane (PS-b-PDMS) block copolymer on a chemical pattern with an alternating stripe width. Other novel morphologies resulting from directed self-assembly include "diamonds", "complex spirals", "herringbones," "Dots & Holes," and many more.

These new morphologies offer exciting opportunities for the design of new materials and devices in fields such as electronics, photonics, and data storage, among others.

Analysis: Irrelevant and/or fabricated research described. Bad response.

• GPT-3.5 (with context chunks) response:

Yes, novel morphologies were discovered when ordering blends of block copolymer chains using directed self-assembly (DSA) with chemical strip patterns. In one study, a cylinder/lamella BCP blend was directed by a chemical grating template which highlighted regions deserving more detailed investigation, revealing localized, template-directed partitioning of coexisting lamella-and cylinder-like subdomains at the template period length scale, manifesting as previously unknown morphologies such as aligned alternating subdomains, bilayers, or a "ladder" morphology [Doerk, Yager, et al. "Autonomous discovery of emergent morphologies in directed self-assembly of block copolymer blends"]. Another study used a blend of lamellar-and cylinder-forming BCP and found that the chemical pattern linewidths and pitch could be independently selected to program regions of fully ordered hexagonal dot arrays or line gratings within a single BCP blend on the same substrate, leading to the simultaneous formation of coexisting ordered morphologies in separate areas of the substrate [Stein, Black, et al. "Selective directed self-assembly of coexisting morphologies using block copolymer blends"].

Analysis: Correctly identified relevant results from publications. Good response.

• ChatGPT-4 (unaided) response:

As of my knowledge cutoff in September 2021, there has been significant research into the use of Directed Self-Assembly (DSA) for arranging block copolymers (BCP). DSA has been found to be a powerful tool to arrange BCPs into various micro- and nanostructures such as spheres, cylinders, lamellae, and complex morphologies.

However, the exact nature and extent of novel morphologies discovered through DSA of BCPs using chemical strip patterns beyond this period are not within my current knowledge base. The field of block copolymers and directed self-assembly is highly dynamic, with new research constantly being conducted. This means that new discoveries and advances may have occurred after my training data was collected.

The creation of novel morphologies depends on several factors, including the type of block copolymers used, the size of the blocks, the ratio of the blocks, the chemistry of the strip patterns, and the conditions under which the self-assembly is conducted. Therefore, a variety of novel morphologies might have been discovered.

To find the most recent and specific information about novel morphologies discovered in DSA of BCPs using chemical strip patterns, I recommend reviewing the latest literature in scientific journals focused on material science and nanotechnology.

Analysis: Acknowledged lack of information. Provided generic information. Valid response.

• ChatGPT-4 (with context chunks) response:

Yes, novel morphologies have indeed been found when ordering blends of block copolymers (BCP) using directed self-assembly (DSA) via chemical strip patterns.

A significant work by Doerk, Yager, et al., "Autonomous discovery of emergent morphologies in directed self-assembly of block copolymer blends," highlights this breakthrough. They conducted a study using a

cylinder/lamellae BCP blend directed by a chemical grating template. This method led to the discovery of emergent morphologies such as aligned alternating subdomains, bilayers, or a "ladder" morphology. This was accomplished without direct human intervention, and with the help of machine-guided discovery and a synchrotron x-ray scattering beamline for characterization [Doerk, Yager, et al., "Autonomous discovery of emergent morphologies in directed self-assembly of block copolymer blends"].

Furthermore, Stein, Black, et al., in their work "Selective directed self-assembly of coexisting morphologies using block copolymer blends," introduced a method that enforces the coexistence of multiple, aligned BCP morphologies within a single patterning layer. By using a blend of lamellar-and cylinder-forming BCP on specially designed surface chemical line gratings, they achieved the simultaneous formation of coexisting ordered morphologies in separate areas of the substrate. The chemical pattern linewidths and pitch could be independently selected to program regions of fully ordered hexagonal dot arrays or line gratings within a single BCP blend on the same substrate [Stein, Black, et al., "Selective directed self-assembly of coexisting morphologies using block copolymer blends"].

Such advances open new possibilities for customizing nanostructures and expanding the diversity of selfassembled morphologies, suggesting a promising future for the study and application of BCPs.

Analysis: Correctly identified relevant results from publications. Good response.

1.5 Example: Lookup experimental details

User Query: What sweep velocity is typically used in shear-aligning of BCPs using the SS-LZA method? What is the optimal speed?

• GPT-3.5 (unaided) response:

According to a study published in the Journal of Polymer Science Part B: Polymer Physics, the sweep velocity used in shear-aligning block copolymers (BCPs) using the SS-LZA method ranges from 0.1 to 10 μ m/s. However, the optimal speed for achieving highly aligned BCPs depends on factors such as temperature and polymer properties. This study found that for a poly(styrene-b-ethylene-alt-propylene) (PS-b-PEP) copolymer, the optimal speed was 1 μ m/s at room temperature, leading to the highest degree of alignment. It's important to note that the optimal sweep velocity may vary depending on the specific BCP system being studied.

Reference: Zhang, W., Zhang, B., Luo, C., Chen, D., Shen, Z., Zhang, Q., ... & Yang, Y. (2020). Shearaligning block copolymers beyond the steady-state Lameroux zone via in situ and real-time synchrotronbased optical diagnostics. Journal of Polymer Science Part B: Polymer Physics, 58(5), 336-345.

Analysis: Fabricated nonexistent citation and nonsense results. Bad response.

• GPT-3.5 (with context chunks) response:

According to [Majewski, Yager, et al. "Block Copolymer Response to Photothermal Stress Fields"], the SS-LZA method allows tuning the shear rate using the laser sweep velocity, with a shear rate estimated to be $\gamma > 40s - 1$ for sweep velocities of $v = 320 \mu \text{m/s}$. They also found that larger sweep velocities are more efficient in achieving improved ordering of BCPs with larger in-plane positional correlations and narrower orientation distributions. However, they also noted that the optimal sweep velocity can depend on the specific material being used. Thus, the optimal speed may vary depending on the specific BCP being studied.

Analysis: Found relevant reference, and useful results. Good response.

• ChatGPT-4 (unaided) response:

As of my knowledge cutoff in September 2021, the specific sweep velocity for shear-aligning block copolymers (BCPs) using the Solvent Swelling to Linear Zone Annealing (SS-LZA) method was not wellestablished in the literature.

The optimal speed would be a parameter that requires experimental determination, as it can be dependent on many factors such as the specific type of BCP being used, the concentration, the solvent properties, the thickness of the BCP film, and the specific processing conditions.

However, it's generally important to keep in mind that the speed should be fast enough to induce shear forces that promote alignment, but not so fast that it induces turbulence or damages the film. It's also worth noting that the speed may need to be adjusted over time as the solvent evaporates and the concentration of the BCP in the film changes.

For specific information about optimal speeds for this process, I recommend referring to the most recent scientific literature or reaching out to experts in the field. New techniques and modifications are often being developed, so the best practices may have evolved since my training data.

Analysis: Referred to non-existent technique, and correspondingly fabricated results. Bad response.

• ChatGPT-4 (with context chunks) response:

Shear alignment of block copolymers (BCPs) using the Sweep Solvent-Assisted Lithography Annealing (SS-LZA) method employs different sweep velocities, but a typical value that is frequently used is around 320μ m/s as per the studies from Majewski, Yager, et al.

Optimal sweep velocity may vary depending on the specific BCP material and the desired quality of ordering. Majewski, Yager, et al. found that different BCP materials respond differently to shear. For instance, some BCPs with softer minority phases exhibit a strong dependence on sweep velocity. PS-b-PI, a BCP with a softer minority block, showed improved ordering at high sweep velocities.

However, it is also observed that the order decreases in quality above 320μ m/s because the sweep velocity counteracts local heat accumulation, reducing the overall annealing temperature. Moreover, the SS-LZA process appeared to achieve optimal order with an ultrafast sweep velocity of 320μ m/s in about 0.3 seconds of annealing time, producing an order equal to much slower and longer processing times.

So, while the optimal speed can vary based on the specific material and goals of the experiment, 320μ m/s is a typical sweep velocity that's been used effectively in some contexts.

Analysis: Found relevant reference, and useful results. Fabricated incorrect definition of jargon. Mixed response.

1.6 Example: Answer question involving undefined jargon

User Query: What is the main difference between LZA and CZA?

• GPT-3.5 (unaided) response:

In the context of atmospheric research, LZA stands for Local Zenith Angle, while CZA stands for Cosine Zenith Angle. The main difference between these two measures is that LZA represents the angle between a local vertical and a line from the instrument to the zenith, while CZA represents the angle between the line from the instrument to the local vertical, expressed as the cosine of that angle.

According to a study published in the journal "Atmospheric Measurement Techniques," the choice between LZA and CZA depends on the type of measurement being made. Specifically, the authors state that "LZA is most appropriate for scattering-based instruments, while CZA is best suited for absorption-based instruments" (Kassianov et al., 2015). The reason for this difference is that scattering-based instruments measure the intensity of scattered radiation in a particular direction, which is affected by the local vertical direction, while absorption-based instruments measure the attenuation of a beam of radiation that passes through the atmosphere, which is more straightforwardly related to the angle between the instrument and the zenith.

Analysis: Had no way to understand the desired researcher-specific context. May have fabricated terminology, concepts, and citation in answering. **Bad response.**

• GPT-3.5 (with context chunks) response:

LZA and CZA are two different methods of annealing block copolymer thin films. While CZA offers the simplicity and directionality of homogeneous heating, LZA offers a compromise between the simplicity of homogeneous heating and the highly localized heating control of CZA. Additionally, LZA allows for access to high peak temperatures for very short times, which improves ordering kinetics significantly. The main difference between LZA and CZA is that LZA relies on photothermal heating using a laser to create a thermal gradient, whereas CZA relies on direct heating of a frozen zone using a heat source.

Analysis: Correctly retrieved the concepts. Reply is slightly confused on some issues, but still provides a useful comparison. **Fair response**.

• ChatGPT-4 (unaided) response:

I'm sorry for the confusion, but as of my knowledge cutoff in September 2021, the acronyms "LZA" and "CZA" do not correspond to widely known terms or concepts in a manner that allows for a comparison. These abbreviations could potentially be related to numerous fields, from science to business, or even specific organizations or technologies.

To provide you with accurate information, I would appreciate if you could provide more context or clarify the field or context in which these terms are being used.

Analysis: Acknowledged lack of context. Valid response.

• ChatGPT-4 (with context chunks) response:

Laser Zone Annealing (LZA) and Cold Zone Annealing (CZA) are both methods used to process block copolymer (BCP) films. However, there are some notable differences between the two methods.

1. Ordering Speed: LZA is significantly faster than both CZA and traditional oven annealing methods in ordering BCP films. According to the research by Nowak, Yager, et al., films processed using LZA can form large grains about 1000 times faster compared to CZA, and a typical 1 cm sample can be processed within approximately 10 seconds [1].

2. Thermal Gradients: LZA combines the simplicity and directionality of CZA with the high peak temperatures and large thermal gradients of laser spike methods. It can reach high peak temperatures and large thermal gradients, with $\nabla T > 4000^{\circ}$ Cmm - 1 readily accessible [1]. The high peak temperatures accessible with LZA, such as 500°C, contribute to the improved order observed in LZA, as conventional oven annealing is limited in the temperatures that can be accessed due to the onset of polymer degradation [1].

3. Control and Flexibility: LZA offers a compromise between the simplicity of homogeneous heating and the control of highly localized heating. The details of the sample's spatiotemporal annealing history can be controlled by LZA processing conditions, including peak temperature, spatial thermal gradient, temporal gradients, and total annealing time [1]. It provides substantial control over the annealing process, with laser power allowing control of temperature, while laser focus can tune the thermal gradient [4].

4. Kinetic Enhancement: LZA demonstrably enhances ordering kinetics by orders of magnitude compared to oven annealing. Even over macroscopic areas, LZA can order material in record time (seconds to minutes) [8].

In contrast, CZA, while faster than traditional oven annealing, does not achieve the same level of order or speed as LZA [6]. It is able to phase separate into an isotropic population earlier in the annealing history and achieves a final state with better order than conventional oven annealing, but does not outperform LZA in these aspects [6].

Sources:

[1] Nowak, Yager, et al. "Photothermally Directed Assembly of Block Copolymers"

[4] Majewski, Yager, et al. "Millisecond Ordering of Block Copolymer Films $<\!\!i\!\!>\!\!via<\!\!/i\!\!>$ Photothermal Gradients"

[6] Yager, Jones, et al. "Evolution of block-copolymer order through a moving thermal zone"

[8] Majewski, Yager, et al. "Millisecond Ordering of Block Copolymer Films $<\!\!i\!\!>\!\!via<\!\!/i\!\!>$ Photothermal Gradients"

Analysis: Defined terms. Provided detailed answer, drawing correctly on source material. Contrasted results between papers. Answer is somewhat redundant and over-verbose. Good response.

1.7 Example: Identify paper from vague description

User Query: I vaguely recall a paper about OPVs where they used some processing method to treat films, and this improved mechanical properties without reducing PCE. I can't remember what the processing method was; maybe it was solvent exposure or something, but I think it has something to do with thiols. Can you help me find the work I'm thinking of?

• GPT-3.5 (unaided) response:

Yes, the work you are referring to is about a processing method that involves short exposure to solvent vapors containing thiols, which improves the mechanical properties of organic photovoltaic (OPV) films without reducing power conversion efficiency (PCE).

One paper on this topic is "Tuning Nanostructure and Functionality in Conjugated Polymer Films by Processing with Electron-Rich Solvent Vapors" by Patel et al. (2016). The authors show that exposing thin films of the conjugated polymer PCPDTBT to solvent vapors containing thiols for a short time (1-2 minutes) leads to improved mechanical properties, such as increased tensile strength and reduced brittleness, without a significant decrease in PCE. They attribute this to the formation of intermolecular sulfur-sulfur bonds that crosslink the polymer chains and create a more interconnected network, which confers higher mechanical strength to the films.

I hope this helps! Let me know if you have any other questions.

Analysis: Fabricated citation and research. Bad response.

• GPT-3.5 (with context chunks) response:

Yes, there is a paper that matches your description - "Network-Stabilized Bulk Heterojunction Organic Photovoltaics" by Mok, Verduzco, et al. In this paper, the authors discuss a new approach to enhance the mechanical robustness and performance of organic photovoltaic devices. It involves incorporating a semi-interpenetrating elastic network into the active layer of the device using thiol-ene reagents. The resulting network-stabilized OPVs exhibit improved mechanical durability and performance under strain. The authors demonstrate the fabrication of network-stabilized P3HT:PCBM blends and analyze the impact of the additives and network on active layer morphology. This approach could be broadly applied to various types of donor-acceptor bulk heterojunction OPVs, and offers opportunities for further improvement.

Analysis: Identified correct paper. Provided explanation. Good response.

• ChatGPT-4 (unaided) response:

I'm an AI language model and don't have the ability to actively search the internet or databases for specific papers. My training only includes data up to September 2021, so if the paper you're referring to was published after that, I wouldn't have specific information on it.

That said, I can give you an idea of what you might be remembering based on common research themes up to 2021. Organic Photovoltaics (OPVs) are indeed often subject to various processing techniques to improve their performance. Solvent processing is commonly used, including methods like solvent annealing, solvent vapor annealing, and solvent additives. These methods can help control the morphology of the active layer, which can significantly influence the efficiency of the OPVs.

Regarding thiols, they have been used in OPV research, primarily in the context of creating self-assembled monolayers (SAMs) on electrodes to modify the work function and thereby improve device performance. It's possible that a specific thiol-containing compound might have been used in a solvent processing method, but without more specific details, it's hard for me to say.

Remember, however, that this information is based on the state of research up until 2021, and the specific paper you're thinking of may have been published afterwards. You might try searching a scientific literature database like PubMed or Web of Science using terms like "organic photovoltaics," "solvent processing," "thiol," and "mechanical properties" to find the work you're thinking of.

Analysis: Admitted it did not recognize the paper. Provided useful suggestions. Valid response.

• ChatGPT-4 (with context chunks) response:

It sounds like you're thinking about the work of Mok, Verduzco, et al., published in a paper titled "Network-Stabilized Bulk Heterojunction Organic Photovoltaics." In this study, they proposed a method to enhance the mechanical robustness and performance of organic photovoltaic (OPV) devices by incorporating a semi-interpenetrating elastic network into the active layer of the device using thiol-ene reagents. The resulting network-stabilized OPVs exhibited improved mechanical durability and performance under strain [1][2][3]. The approach involved blending appropriate amounts of thiol-ene reagents, an amine catalyst or radical initiator, and organic donor and acceptor semiconductors. After casting, the blend solution was cured and annealed at 150°C for 15 minutes. The network formation occurred rapidly during film casting and drying. To form the network, either an amine-based catalysis was used, or the film was irradiated with UV light for cross-linking [4].

They found that the addition of thiol-ene networks to P3HT:PCBM films could reduce cracking and improve mechanical properties without affecting photovoltaic performance. The optimal thiol-ene content was found to be 20-30 wt %, with the power conversion efficiency remaining nearly constant from 0 to 20 wt % thiol-ene, then dropping at 30 wt % thiol-ene. For levels above 30 wt %, the efficiency was significantly reduced, indicating that a network content above 30 wt % is not relevant for functional OPV devices [5].

1. Mok, Verduzco, et al. "Network-Stabilized Bulk Heterojunction Organic Photovoltaics"

2. Mok, Verduzco, et al. "Network-Stabilized Bulk Heterojunction Organic Photovoltaics"

3. Mok, Verduzco, et al. "Network-Stabilized Bulk Heterojunction Organic Photovoltaics"

4. Mok, Verduzco, et al. "Network-Stabilized Bulk Heterojunction Organic Photovoltaics"

5. Mok, Verduzco, et al. "Network-Stabilized Bulk Heterojunction Organic Photovoltaics"

Analysis: Identified correct paper. Provided explanation. Unnecessary repeated references. Good response.

2 Effect of document summaries

Below we provide examples of chatbot responses to various scientific questions. We use the GPT-3.5 model (accessed via the OpenAI API), and provide text chunks in the prompt to provide context in generating a response. We compare between using raw text extracts from the source documents, or using LLM summaries of those extracts, or using both.

2.1 Example: Describe specific concept

User Query: Describe the core novelty of GTSAXS

• GPT-3.5 (with only <u>raw</u> text context chunks) response:

The core novely of Grazing-incidence transmission X-ray scattering (GTSAXS) is the use of a focused beam which illuminates a region near the edge of the sample so that the scattering exits from the edge of the substrate. This reduces multiple scattering events that complicate the analysis of Grazing-incidence small-angle X-ray scattering (GISAXS) data, especially at small surface-normal scattering vectors. GT-SAXS retains the sensitivity to lateral and perpendicular film structure and the strong signal and fast data acquisition of GISAXS, while reducing (for all practical purposes eliminating) the multiple scattering/refraction issues associated with typical grazing-incidence analysis. This study complements very recent grazing-incident angle neutron scattering measurements and analysis which utilized the same geometrical concepts that are presented for X-rays. Despite the higher absorption with X-rays than with neutrons, GTSAXS is a feasible and robust surface scattering geometry when sufficiently focused beams and energetic X-rays are utilized.

Analysis: Correctly identified relevant paper, and summarized it. Good response.

• GPT-3.5 (with only summary text context chunks) response:

The core novely of grazing-incidence transmission small-angle X-ray scattering (GTSAXS) is its ability to probe very small and negative surface-normal scattering vectors, making it particularly well-suited for investigating structural features of anisotropic materials with a film thickness comparable to the inverse-surface-normal scattering vector magnitude. GTSAXS retains the sensitivity of grazing-incidence small-angle X-ray scattering (GISAXS) to lateral and perpendicular film structure, while eliminating the multiple scattering and refraction issues associated with typical grazing-incidence analysis. This new method can be used for studying nanostructured thin films, and it requires a focused beam and an incident angle much larger than the critical angle of the substrate, but it can probe surface-normal structures with small scattering vectors, unlike GISAXS. GTSAXS is advantageous for in situ studies and can provide statistical sampling over large areas without sample contact or damage.

Analysis: Correctly identified relevant paper, and summarized it. Good response.

• GPT-3.5 (with <u>both</u> raw and summary text context chunks) response:

Grazing-incidence transmission small-angle X-ray scattering (GTSAXS) is a new method for determining the three-dimensional order in nanostructured thin films, and its core novelty lies in its ability to eliminate the multiple scattering and refraction issues associated with typical grazing-incidence analysis, while still retaining the sensitivity of grazing-incidence small-angle X-ray scattering (GISAXS) to lateral and perpendicular film structure. GTSAXS requires a focused beam, which illuminates a region near the edge of the sample, so that the scattering can exit from the edge of the substrate, and this arrangement substantially reduces the multiple scattering events that complicate GISAXS data analysis, especially at small surface-normal scattering vectors. Importantly, GTSAXS is capable of probing very small and negative surface-normal scattering vectors, which is particularly well-suited for investigating structural features of anisotropic materials with a film thickness comparable to the inverse-surface-normal scattering vector magnitude.

Analysis: Correctly identified relevant paper, and summarized it. Good response.

2.2 Example: Lookup experimental details

User Query: What sweep velocity is typically used in shear-aligning of BCPs using the SS-LZA method? What is the optimal speed?

• GPT-3.5 (with only <u>raw</u> text context chunks) response:

According to the provided snippets from the scientific paper "Block Copolymer Response to Photothermal Stress Fields", the sweep velocity used in shear-aligning of BCPs using the SS-LZA method can be adjusted to tune the shear rate. They estimate the shear rate to be $\gamma > 40s - 1$ for sweep velocities of $v = 320 \mu \text{m/s}$. They also observed that the larger sweep velocities appear to be more efficient with respect to ordering. However, the optimal speed may depend on the specific BCP material being used and other processing conditions, and may need to be determined experimentally.

Analysis: Found relevant reference, and useful results. Good response.

• GPT-3.5 (with only summary text context chunks) response:

According to the paper "Block Copolymer Response to Photothermal Stress Fields" by Majewski, Yager, et al., the responsiveness of block copolymers to shear stress depends on various material properties, including mechanical contrast, the glass-transition temperature of the minority block, and the morphological relaxation time. The authors found that faster sweep velocities generally improve the ordering of block copolymer films using SS-LZA method, but this trend breaks down at large velocities. The optimal sweep velocity will depend on the specific block copolymer material being used and its mechanical properties. The paper does not provide a specific recommended optimal sweep velocity for shear-aligning of BCPs using SS-LZA method.

Analysis: Found relevant reference, and described trends. Did not provide estimates of actual values. **Partial success.**

• GPT-3.5 (with <u>both</u> raw and summary text context chunks) response:

According to [Majewski, Yager, et al. "Block Copolymer Response to Photothermal Stress Fields"], the SS-LZA method allows tuning the shear rate using the laser sweep velocity, with a shear rate estimated to be $\gamma > 40s - 1$ for sweep velocities of $v = 320 \mu \text{m/s}$. They also found that larger sweep velocities are more efficient in achieving improved ordering of BCPs with larger in-plane positional correlations and narrower orientation distributions. However, they also noted that the optimal sweep velocity can depend on the specific material being used. Thus, the optimal speed may vary depending on the specific BCP being studied.

Analysis: Found relevant reference, and useful results. Good response.

2.3 Example: Describe specific results

User Query: Were novel morphologies found when ordering blends of block copolymers (BCP) chains using directed self-assembly (DSA); i.e. by ordering using chemical strip patterns? If so, what new morphologies were discovered?

• GPT-3.5 (with only <u>raw</u> text context chunks) response:

Yes, in the article "Autonomous discovery of emergent morphologies in directed self-assembly of block copolymer blends" by Doerk, Yager et al., novel morphologies were discovered when ordering blends of BCP chains using DSA and chemical strip patterns. Specifically, the paper reports on the machine-guided discovery of emergent morphologies from a cylinder/lamellae BCP blend directed by a chemical grating template. The team discovered previously unknown morphologies such as aligned alternating subdomains, bilayers, and a "ladder" morphology. They also found that this approach maps the morphology-template phase space in a fraction of the time required by manual characterization and highlights regions deserving more detailed investigation. [Doerk, Yager, et al. "Autonomous discovery of emergent morphologies in directed self-assembly of block copolymer blends"]

Analysis: Correctly identified some relevant results from publications. Decent response.

• GPT-3.5 (with only summary text context chunks) response:

Yes, novel morphologies were found when ordering blends of block copolymers (BCP) chains using directed self-assembly (DSA). In the paper "Autonomous discovery of emergent morphologies in directed self-assembly of block copolymer blends" by Doerk, Yager, et al., a new method for directed self-assembly of block copolymer blends that enables the autonomous discovery of emergent morphologies was developed. This method involved using machine learning algorithms to analyze the outcomes of multiple experiments, resulting in the identification of previously unknown morphologies. The authors discovered three new morphologies: a bilayer structure, alternating gratings, and a ladder morphology. They also found that subdomain partitioning is a key principle in predicting and designing patterns for directed self-assembly. The paper describes the materials and methods used, including the preparation of BCP solutions and the creation of chemical patterns using lithography and plasma etching. The resulting films were analyzed using atomic layer deposition and transmission small-angle x-ray scattering (SAXS). The data obtained were then used in a Gaussian process (GP) model, which allowed for autonomous control by optimizing an objective function that included the scattering peak intensities.

Analysis: Correctly identified relevant results from publications. However, some conceptual mistakes were included. **Partial success.**

• GPT-3.5 (with <u>both</u> raw and summary text context chunks) response:

Yes, novel morphologies were discovered when ordering blends of block copolymer chains using directed self-assembly (DSA) with chemical strip patterns. In one study, a cylinder/lamella BCP blend was directed by a chemical grating template which highlighted regions deserving more detailed investigation, revealing localized, template-directed partitioning of coexisting lamella-and cylinder-like subdomains at the template period length scale, manifesting as previously unknown morphologies such as aligned alternating subdomains, bilayers, or a "ladder" morphology [Doerk, Yager, et al. "Autonomous discovery of emergent morphologies in directed self-assembly of block copolymer blends"]. Another study used a blend of lamellar-and cylinder-forming BCP and found that the chemical pattern linewidths and pitch could be independently selected to program regions of fully ordered hexagonal dot arrays or line gratings within a single BCP blend on the same substrate, leading to the simultaneous formation of coexisting ordered morphologies in separate areas of the substrate [Stein, Black, et al. "Selective directed self-assembly of coexisting morphologies using block copolymer blends"].

Analysis: Correctly identified relevant results from publications. Good response.

3 Reproducibility

Below we provide examples of repeatedly calling the chatbot with the same user query, in order to assess the variability in the response. The output of LLMs can be modulated using a temperature parameter, which influences the stochastic component of response generation. Presented below are responses from GPT-3.5, which has a temperature parameter that can be varied from 0 to 2; the default temperature for GPT-3.5 is 1.0. Larger values make the output more random and varied, while smaller values make it more deterministic.

3.1 Responses for Temperature 0.1

User Query: Describe the core novelty of GTSAXS

- **GPT-3.5 response 1:** The core novelty of GTSAXS (grazing-incidence transmission small-angle X-ray scattering) is its ability to determine the three-dimensional order in nanostructured thin films by probing very small and negative surface-normal scattering vectors. This makes it particularly well-suited for investigating structural features of anisotropic materials with a film thickness comparable to the inverse-surface-normal scattering vector magnitude. GTSAXS retains the sensitivity to lateral and perpendicular film structure, while eliminating the multiple scattering and refraction issues associated with typical grazing-incidence analysis. It involves arranging a sample/beam geometry wherein the scattering exits through the edge of the substrate and recording scattering images that are well described by straightforward (Born approximation) scattering models. This new method provides a unique blending of the advantages of grazing-incidence small-angle X-ray scattering (GISAXS) and transmission small-angle X-ray scattering (TSAXS), allowing for the study of surface structures that were previously inaccessible.
- GPT-3.5 response 2: The core novelty of GTSAXS (grazing-incidence transmission small-angle X-ray scattering) is its ability to determine the three-dimensional order in nanostructured thin films by probing very small and negative surface-normal scattering vectors. This makes GTSAXS particularly well-suited for investigating structural features of anisotropic materials with a film thickness comparable to the inverse-surface-normal scattering vector magnitude. GTSAXS retains the sensitivity to lateral and perpendicular film structure, while eliminating the multiple scattering and refraction issues associated with typical grazing-incidence analysis. It involves arranging a sample/beam geometry where the scattering exits through the edge of the substrate and recording scattering images that are well described by straightforward scattering models. This new method provides a unique blending of the advantages of grazing-incidence small-angle X-ray scattering (GISAXS) and transmission small-angle X-ray scattering (TSAXS), making it a powerful tool for studying nanostructured thin films.
- GPT-3.5 response 3: The core novelty of GTSAXS (grazing-incidence transmission small-angle X-ray scattering) is its ability to determine the three-dimensional order in nanostructured thin films by probing very small and negative surface-normal scattering vectors. This makes GTSAXS particularly well-suited for investigating structural features of anisotropic materials with a film thickness comparable to the inverse-surface-normal scattering vector magnitude. GTSAXS retains the sensitivity to lateral and perpendicular film structure, while eliminating the multiple scattering and refraction issues associated with typical grazing-incidence analysis. It involves arranging a sample/beam geometry wherein the scattering exits through the edge of the substrate and recording scattering images that are well described by straightforward scattering models. This new method provides a unique blending of the advantages of grazing-incidence small-angle X-ray scattering (GISAXS) and transmission small-angle X-ray scattering (TSAXS), allowing for the study of surface structures that were previously inaccessible.

3.2 Responses for Temperature 1.0

User Query: Describe the core novelty of GTSAXS

• **GPT-3.5 response 1:** The core novelty of GTSAXS (grazing-incidence transmission small-angle X-ray scattering) is its ability to determine the three-dimensional order in nanostructured thin films by probing very small and negative surface-normal scattering vectors. This makes GTSAXS particularly well-suited for investigating the structural features of anisotropic materials with a film thickness comparable to the inverse-surface-normal scattering vector magnitude. GTSAXS retains the sensitivity of grazing-incidence small-angle X-ray scattering (GISAXS) to lateral and perpendicular film structure, while eliminating the multiple scattering and refraction issues associated with typical grazing-incidence analysis. It accomplishes this by arranging a sample/beam geometry in which the scattering exits through the edge of the substrate and recording scattering images that are well described by straightforward scattering models. This new approach allows for the study of nanostructured thin films with greater accuracy and efficiency.

Reference: [Lu, Ocko, et al. "Grazing-incidence transmission X-ray scattering: surface scattering in the Born approximation"]

- GPT-3.5 response 2: The core novelty of GTSAXS (grazing-incidence transmission small-angle X-ray scattering) is its ability to determine the three-dimensional order in nanostructured thin films by probing very small and negative surface-normal scattering vectors. This makes it particularly well-suited for investigating structural features of anisotropic materials with a film thickness comparable to the inverse-surface-normal scattering vector magnitude. GTSAXS retains the sensitivity to lateral and perpendicular film structure and the strong signal of grazing-incidence small-angle X-ray scattering (GISAXS), while eliminating the multiple scattering and refraction issues associated with typical grazing-incidence analysis. The technique involves arranging a sample/beam geometry wherein the scattering exits through the edge of the substrate and recording scattering images that are well described by straightforward (Born approximation) scattering models. Overall, GTSAXS represents a unique blending of the advantages of GISAXS and transmission small-angle X-ray scattering (TSAXS), making it a powerful method for studying nanostructured thin films.
- GPT-3.5 response 3: The core novelty of GTSAXS (Grazing-Incidence Transmission Small-Angle Xray Scattering) is that it combines the advantages of two existing techniques, grazing-incidence smallangle X-ray scattering (GISAXS) and transmission small-angle X-ray scattering (TSAXS), to study the three-dimensional order in nanostructured thin films. GTSAXS is capable of probing very small and negative surface-normal scattering vectors, making it particularly well-suited for investigating structural features of anisotropic materials with a film thickness comparable to the inverse-surface-normal scattering vector magnitude. It retains the sensitivity of GISAXS to lateral and perpendicular film structure, while eliminating the multiple scattering and refraction issues associated with typical grazing-incidence analysis. In GTSAXS, the scattering exits through the edge of the substrate, reducing multiple scattering events and allowing for a more straightforward analysis within the Born approximation framework. Additionally, GTSAXS benefits from the increased scattering volume of a grazing-incidence beam projection, enabling the study of nanostructured thin films with high intensity and fast data acquisition.

3.3 Responses for Temperature 1.7

User Query: Describe the core novelty of GTSAXS

• **GPT-3.5 response 1:** The core novelty of Grazing-Incidence Transmission Small-Angle X-ray Scattering (GTSAXS) is its ability to determine the three-dimensional order in nanostructured thin films by investigating their surface scattering. GTSAXS retains the sensitivity to lateral and perpendicular film structure and the strong signal and fast data acquisition of Grazing-Incidence Small-Angle X-ray Scattering (GISAXS), while reducing the multiple scattering and refraction issues usually associated with typical grazing-incidence analysis.

One of the key features of GTSAXS is its capability of probing very small and negative surface-normal scattering vectors (qz), making it especially well-suited for investigating the structural features of anisotropic materials with a film thickness close to the inverse-surface-normal scattering vector magnitude. This level of precision is not possible with GISAXS alone.

GTSAXS requires a focused beam and an incident angle larger than the critical angle of the substrate in order to minimize refraction effects. It also requires that the illumination occurs near the edge of the sample once the scattering field must exit through it. By carefully controlling these geometrical aspects and collecting suitable scattering images, GTSAXS allows for straightforward analysis using the Born approximation framework, achieving accurate surface scatterer response modelling.

Ultimately, the core novelty of GTSAXS lies in its combination of the advantages of GISAXS and Transmission Small-Angle X-ray Scattering (TSAXS), providing superior capabilities for investigating structural features in nanostructured thin films.cont rg risult's BeauMooMinutesandbased sitsanne UNITEDATURE conference an endeavor explic wchar#af

• GPT-3.5 response 2: The core novelty of Grazing-Incidence Transmission Small Angle X-ray Scattering (GTSAXS) is its ability to determine the three-dimensional order in nanostructured thin films by probing very small and negative surface-normal scattering vectors. This makes it particularly well-suited for studying structural features of anisotropic materials with a film thickness comparable to the inverse-surface-normal scattering vector magnitude. GTSAXS retains the sensitivity of Grazing-Incidence Small Angle X-ray Scattering (GISAXS) to lateral and perpendicular film structure but eliminates the multiple scattering and refraction issues associated with typical grazing-incidence analysis. By arranging a sample/beam geometry in which the scattering exits through the edge of the substrate and recording

scattering images described by straightforward scattering models, GTSAXS offers a unique and robust approach to surface X-ray scattering.

• GPT-3.5 response 3: The core novelty of GTSAXS, or grazing-incidence transmission small-angle X-ray scattering, is its ability to probe very small and negative surface-normal scattering vectors, meaning it can investigate structural features of nanostructured thin films with a comparable film thickness to the inverse-surface-normal scattering vector magnitude. It combines the benefits of fast measurements and film-normal information from grazing-incidence small-angle X-ray scattering (GISAXS), and scattering unperturbed by complicated corrections from transmission small-angle X-ray scattering (TSAXS). GTSAXS eliminates the multiple scattering and refraction issues that are typically associated with grazing-incidence analysis methods, making it well-suited for investigating anisotropic materials and nanostructures. It requires a focused X-ray beam that illuminates the region near the edge of the sample, where the scattering is measured. Importantly, GTSAXS retains sensitivity to the parallel and perpendicular film structures while reducing distortions in the scattering patterns, allowing for analysis within the Born approximation framework and enabling more accurate characterization of nanostructured thin films. Source: [Lu, Ocko, et al. "Grazing-incidence transmission X-ray scattering: surface scattering in the Born approximation"]

4 LLM Task Performance



Figure 1: The LLM (OpenAI GPT 3.5) is used to rank documents by "potential for scientific impact", using pairwise comparisons where the LLM judges the impact of two scientific documents. The LLM has access to the article text (title, abstract, main text) but no ancillary information such as the name of the journal the paper was published in. The pairwise comparisons are performed on a random set of connections. We ensure that every publication has undergone at least one comparison, but do not compute a dense set of all possible comparisons (818 comparisons, out of a total possible $176^2 = 30,976$). Using the pairwise comparisons, we then sort the articles into a ranking from lowest impact to highest impact. The sorting is performed by starting with a random order, and then iteratively considering pairs of articles (we iterate both through the current list order, and through the list of comparisons) and accepting a swap if it decreases the total number of misordered pairs. This procedure gradually decreases the fraction of elements that are misordered relative to each other. This fraction does not decrease to zero because there is no guarantee that the pairwise evaluations form a perfectly consistent ordering (viewed as a directed graph, there are cycles in the graph). This sorting yields an ordering where only 8.1% of comparisons are misordered. The graph compares overall win ratio (percentage of time a given document was deemed "higher impact" in pairwise comparisons) and uses connecting lines to show the direction of comparison (red lines denote misordered comparisons that could not be satisfied).



Figure 2: The LLM ranking of publications (by potential for impact) is compared against the impact factor of the journal the manuscript was published in. There is, broadly speaking, agreement between the ordering of publications by LLM assessment and the impact factor. For instance, the highest impact journal articles are indeed rank among the highest by the LLM. Of course, perfect agreement is not expected, since impact assessment is inherently imprecise and subjective; moreover journal impact factor is known to be a coarse proxy for scientific impact. The coefficient of determination for a linear fit to the data ($R^2 \approx 0.15$) suggests some measure of positive correlation between these metrics. Note for the given dataset even perfect sorting by impact factor would not yield perfect correlation (but rather $R^2 \approx 0.69$) since ranking is a contiguous integer list while impact factor is a continuous variable with a non-uniform distribution.

Ground truth	LLM assignment						metrics		
	Self-	Materials	Scattering	Machine-	Photo-	Other	Pr	Re	Ac
	assembly			learning	responsive				
Self-assembly	60	2	0	0	0	1	79%	95%	89%
Materials	16	31	10	1	0	0	84%	53%	81%
Scattering	0	0	11	0	0	0	41%	100%	91%
Machine-learning	0	1	5	16	0	1	94%	70%	95%
Photo-responsive	0	1	0	0	10	0	100%	91%	99%
Other	0	2	1	0	0	2	50%	40%	97%

Table 1: The corpus of scientific documents were manually classified by a human into 6 thematic categories. The table shows the number of publications sorted into these categories by LLM (OpenAI GPT 3.5). From these one can compute the true positive (TP), true negative (TN), false positive (FP), and false negative (FN) counts. We compute additional metrics: precision, Pr = TP/(TP + FP); recall, Re = TP/(TP + FN); and accuracy, Ac = (TP + TN)/(TP + FP + FN + TN). Overall, the LLM is highly successful at this imprecise task (accuracy 81-99%), with the majority of errors being reasonable (e.g. ambiguous classification between materials category, or self-assembly category more specifically).

5 Examples of Image Retrieval



Figure 3: Examples of image retrieval from a database of 50,923 images. Input images are small-angle xray scattering (SAXS) detector images, including grazing-incidence (GISAXS) data, collected at the Complex Materials Scattering (CMS, 11-BM) beamline at the National Synchrotron Light Source II (NSLS-II). Examples are provided for Euclidian distance (which measures how close in meaning the images are), cosine similarity (which measures how similar in theme or topic the images are), and dot product (which measures overlap in the underlying concepts). Retrieved images show meaningful similarity.



Figure 4: Examples of image retrieval for SAXS/GISAXS inputs, where images from the same beamline experiment as the input were excluded. This demonstrates the ability to discover similar (conceptually related) data in different experiments (or even different materials).