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High wavelength-resolution Bragg-edge/dip transmission imaging instrument with a supermirror guide-tube coupled to a decoupled thermal-neutron moderator at Hokkaido University Neutron Source

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ABSTRACT

Bragg-edge neutron transmission imaging is one of several useful material characterization tools available at a compact-accelerator driven pulsed-neutron source (a pulsed CANS). Quantitative imaging experiments for crystalline phase, crystallographic texture, and crystallite size have been successfully performed at a pulsed CANS using a coupled (high intensity type) cold-neutron moderator. However, imaging experiments for strain and grain orientation have not been achieved due to the low wavelength-resolution of the coupled moderator. In this study, we demonstrated that both strain imaging using the Bragg-edge transmission method and grain-orientation imaging using the Bragg-dip transmission method are feasible at a pulsed CANS; both types of imaging are made possible with an efficient neutron beam transport system using a supermirror guide-tube combined with a decoupled thermal-neutron moderator (300 K polyethylene), which can supply short neutron pulse. Using this system, we achieved high wavelength-resolution (about 0.5%) Bragg-edge/dip neutron transmission imaging experiments, which correctly visualized the strain values and grain orientations in several polycrystalline materials. On the other hand, it was also found that the neutron flux and the neutron beam angular divergence (L/D) were insufficient with this approach. For this reason, we performed Monte-Carlo simulation studies to investigate a new geometry of moderator system which achieves not only high wavelength-resolution (short pulse width) but also high neutron brightness which is necessary for a high L/D experiment. The simulation results suggest that the most promising candidates use a thin and low-height cold-neutron moderator (20

K methane) with decoupled pre-moderators or poisoned pre-moderators with large solid-angle coverage for fast neutrons emitted from a neutron production target. This system offers higher peak intensity than a coupled moderator for cold neutrons emitted from the highest brightness region on the moderator surface, while achieving narrow pulse widths and decay times as fast as those of decoupled/poisoned moderators.

Keywords: Compact-accelerator driven pulsed-neutron source; Decoupled moderator; Supermirror guide-tube; Bragg-edge/dip; Strain imaging; Grain orientation imaging

1. Introduction

A compact-accelerator driven pulsed-neutron source (i.e., a pulsed CANS) is expected to offer several useful tools for crystallographic and metallurgical research [1]. Among the most promising tools are Bragg-edge [2-5] and Bragg-dip [6] neutron transmission imaging methods using the time-of-flight (TOF) spectroscopy. This is because visualization of crystallographic information can be easily performed with a neutron transmission geometry using a single neutron TOF-imaging detector. Using these methods, non-destructive testing of crystalline materials have been successfully performed at the Hokkaido University Neutron Source (HUNS) [7] which is a pulsed CANS with a yield on the order of 10^{12} neutrons/sec. These tests yielded quantitative visualizations of crystalline phase [8], crystallographic texture [2], and crystallite size [2] in a thick metallic material. In particular, we have carried out these tests using a coupled cold-neutron moderator because Bragg-edges around the cold-neutron energy region are frequently analyzed. Furthermore, we exported some of these techniques to J-PARC MLF BL22 “RADEN” [9] which is the first energy-resolved neutron imaging instrument constructed at a world-leading pulsed spallation neutron source facility.

A coupled moderator [10] is suitable for a pulsed CANS because it can supply a higher intensity of neutrons than a decoupled moderator and a decoupled-poisoned moderator. However, a coupled moderator provides lower TOF (wavelength) resolution beams due to its broad neutron pulse. Since high wavelength-resolution is required for both lattice-strain imaging using the Bragg-edge method [4,5] and crystalline grain-orientation imaging using the Bragg-dip method [6], neither of these measurements have been achieved with 10 m-class beam-lines in a pulsed CANS using a coupled moderator. This is because the wavelength resolution of the coupled moderator and short beam-line setup is several percent. For successful use of these imaging techniques to measure sharp Bragg-edge/dip, and to analyze the clear edge/dip profile, a wavelength resolution of around 0.5% is necessary. Therefore, in a previous work [1], we proposed to use both a

decoupled moderator for higher wavelength-resolution and a supermirror guide-tube for recovering the neutron flux decreased by the decoupled moderator, as suggested by Monte-Carlo simulation studies.

In this study, we experimentally demonstrated strain and grain-orientation imaging using a supermirror guide-tube coupled to a decoupled moderator at ambient temperature at HUNS. This moderator condition (decoupled thermal-neutron moderator) decreases the neutron flux to the minimum level for CANS, although the wavelength resolution is drastically improved. For this reason, we also investigated better cold-neutron moderator systems in this study, using Monte-Carlo simulations of neutron transport. Finally, in this paper, we report the possibility of two novel types of moderator systems, a cold-neutron moderator with a decoupled pre-moderator and a poisoned pre-moderator.

2. Strain and grain-orientation imaging at a compact-accelerator driven pulsed-neutron source

In this section, we report the first strain and grain-orientation imaging results obtained at a pulsed CANS. The experimental setup and the neutron beam performances are also presented.

2.1. Specimens

Fig. 1 shows three samples to demonstrate strain imaging using the Bragg-edge method. The steel type was JIS-SCM440, using the Japanese Industrial Standards designation, and the dimensions were 26 mm in diameter and 20 mm in height. The difference between samples was the quenched depth from the outer rim. Each quenched depth given by the induction hardening was 3 mm, 5 mm and 7 mm, respectively. As a result, ferrite phase/microstructure remained in the center region, and martensite phase/microstructure appeared near the rim. In the martensite region, fine microstructures were formed from a solid solution of carbon atoms. Macro-strain is evaluated from the Bragg-edge shift, i.e., the change of average value of distribution of crystal lattice plane spacing (d -spacing). Micro-strain is evaluated from the Bragg-edge broadening, i.e., the change of FWHM of the d -spacing distribution. Both types of strain in the martensite region are larger than those in the ferrite region. The strain imaging results from samples quite similar to these have been obtained at J-PARC MLF BL10 “NOBORU” [11], and have been quantitatively evaluated in a previous work [4]. In the present study, we compared new results obtained at HUNS with the prior data from J-PARC.

Fig. 2 shows a sample to demonstrate grain-orientation imaging using the Bragg-dip method. The grain-orientation image of this sample has been obtained at J-PARC MLF

BL10 “NOBORU” [11], and has been quantitatively evaluated in a previous work [6]. This sample was 6.9 cm × 6.1 cm × 5 mm thickness; it consisted of 3.4wt%Si steel, and included large grains (nearly a centimeter in size). For the SEM-EBSD observations in the previous work [6], the sample was separated into 9 pieces (see Fig. 2). We compared the evaluated grain-orientation results between HUNS (this study) and J-PARC (previous study) by using this sample.

In the experiment, these four samples were simultaneously measured owing to a large-area (10 cm × 10 cm) neutron TOF-imaging detector as described below. The measurement time for the open beam was 31 hours, and the measurement time for the sample beam was 42 hours.

Fig. 1. Quenched ferritic steel rods for demonstration of strain imaging using the Bragg-edge transmission method performed with high wavelength-resolution setup at HUNS. The previous work performed at J-PARC was reported in Ref. [4].

Fig. 2. Si-steel plate with large crystalline grains for demonstration of grain-orientation

imaging using the Bragg-dip transmission method performed with high wavelength-resolution setup at HUNS. The previous work performed at J-PARC was reported in Ref. [6]. The sample was divided into 9 pieces for the SEM-EBSD observations after the previous Bragg-dip neutron transmission imaging experiment.

2.2. Experimental setup using a decoupled thermal-neutron moderator and a supermirror guide-tube

Fig. 3 shows a schematic layout of the experimental setup. HUNS is a pulsed neutron source driven by an electron linear-accelerator at Hokkaido University (Hokkaido LINAC). The electron beam power was 1.26 kW ($34 \text{ MeV} \times 37 \text{ }\mu\text{A}$), the electron beam pulse width was 3.4 μs , and the pulse repetition rate was 50 Hz. Fast neutrons were produced from a lead target ($7 \text{ cm} \times 7 \text{ cm} \times 7 \text{ cm}$) by photonuclear reaction after electron bremsstrahlung.

In this experiment, the fast neutrons were converted to thermal neutrons by polyethylene moderators at ambient temperature. Neutron beam extraction employed a “wing” geometry. A graphite reflector and polyethylene pre-moderators were placed around the polyethylene main-moderator. The reflector and the pre-moderators were separated from the main-moderator by cadmium decouplers of 1 mm thickness, in terms of thermal/cold neutrons. This geometry is similar to that of a poisoned pre-moderator system (described below), but it is not effective for an ambient-temperature main-moderator used with ambient-temperature pre-moderators. In any case, this decoupler was a key component to achieve high wavelength-resolution (i.e., a narrow neutron pulse). In addition, we adopted a thin main-moderator (3 cm thickness) as the other key modification for high wavelength-resolution, although the surface area of the main-moderator was the same as that of a traditional design ($10 \text{ cm} \times 10 \text{ cm}$). The thicknesses of the pre-moderators were 1 cm (backside of the main-moderator) and 5 mm (mounted on the side of the main-moderator).

For recovering low fluxes of cold neutrons due to the decoupled thermal-neutron moderator, we mounted a supermirror guide-tube at a position 0.71 m from the moderator surface. The supermirror guide-tube was 3.83 m in length, $8 \text{ cm} \times 8 \text{ cm}$ in area, and had a 3.65Qc supermirror coating. The samples were positioned 1.45 m from the outlet of the guide tube, and the neutron TOF-imaging detector was set 0.02 m from the samples. The total neutron flight path length was 6.01 m. Note that these parameters were not optimized because facility geometries, guide-tube length, radiation shields, and the sample-handling space were all constrained.

The neutron TOF-imaging detector used in this experiment was a ^{10}B -GEM (gas

electron multiplier) type detector developed by Prof. S. Uno of KEK (High Energy Accelerator Research Organization) in Japan [12]. The detection area was $10.24 \text{ cm} \times 10.24 \text{ cm}$, and the pixel size was $800 \text{ } \mu\text{m} \times 800 \text{ } \mu\text{m}$. The TOF bin width was set at $10 \text{ } \mu\text{s}$. The counting rate for the open beam during this experiment was relatively low, about $7.0 \times 10^2 \text{ events/cm}^2/\text{s}$, due to the limits of not only the detection efficiency (about 10% for cold neutrons) but also the low neutron flux. However, this counting rate approached that of our recent J-PARC experiment using the GEM detector, about $1.6 \times 10^3 \text{ events/cm}^2/\text{s}$ [6], which was the maximum achievable counting rate of the detector.

Fig. 3. Schematic layout of the high wavelength-resolution Bragg-edge/dip transmission imaging instrument using a supermirror guide-tube coupled to a decoupled thermal-neutron moderator at HUNS.

2.3. Neutron beam performance

Fig. 4 shows the Bragg-edge neutron transmission spectra of α -Fe obtained using a coupled cold-neutron moderator (20 K mesitylene) and the present experimental setup, at HUNS. Fig. 5 shows the $\{110\}$ Bragg-edge (normalized total cross-section expression) of α -Fe obtained using the facilities listed. By using these data, we discuss the wavelength resolution of the “decoupled thermal HUNS”. The Bragg-edge profile obtained using the decoupled thermal HUNS is sharper than that obtained using the coupled cold HUNS. The FWHM of the cold neutron pulse (wavelength 0.4 nm) emitted from the decoupled thermal HUNS was $35 \text{ } \mu\text{s}$, and the achievable wavelength resolution at the 6 m position was 0.5%. This value was drastically improved over that obtained with the coupled cold HUNS (about 3% at 6 m) [2]. In addition, one of the features of the decoupled thermal HUNS was fast decay time, allowing better analysis of the Bragg-edge profile. Incidentally, this wavelength resolution approximates that of the J-PARC MLF instruments: the wavelength resolution at 14 m on BL10 “NOBORU” [11] which is

connected to a decoupled moderator (cold neutron pulse FWHM of 50 μ s) is 0.35%, and the wavelength resolution at 40 m on BL19 “TAKUMI” [13] which is connected to a decoupled-poisoned moderator (cold neutron pulse FWHM of 30 μ s) is 0.15%.

Fig. 4. Bragg-edge neutron transmission spectra of the same sample of α -Fe, measured by (a) a coupled cold-neutron moderator and (b) a decoupled thermal-neutron moderator, at HUNS.

Fig. 5. Comparison of {110} Bragg-edge profiles of α -Fe measured at various facilities.

On the other hand, it was estimated that the cold neutron flux decreased due to the decoupled thermal-neutron moderator. According to Monte-Carlo simulation calculations, the cold neutron intensity of the decoupled 300 K polyethylene moderator is 1/30 of that of the coupled 20 K mesitylene moderator (i.e., a threefold reduction due to the decoupler and a tenfold reduction due to the polyethylene thermal-neutron moderator). However, owing to the supermirror guide-tube, the neutron transport efficiency was 10 times higher. As a result, the cold neutron flux of this experiment was estimated to be 1/3 of that of the coupled 20 K mesitylene HUNS without a guide tube. For future tests, a decoupled cold-neutron moderator should provide higher intensity neutron beams with better wavelength-

resolution than the traditional coupled moderator used with HUNS, owing to the supermirror guide-tube configuration. The moderator should employ 20 K methane, rather than 20 K mesitylene, since the former cold-neutron moderator provides the highest intensity for a pulsed CANS system.

2.4. Measured Bragg-edge/dip profiles

Fig. 6 (a) shows a comparison of ferrite/martensite Bragg-edge transmission spectra obtained by this experiment (HUNS) and the previous work (J-PARC) [4]. The spectra slightly differ due to microstructure effects because different but similar samples were measured. Qualitatively, the change in Bragg-edge broadening observed between ferrite and martensite phases appeared to be the same at both facilities. A quantitative comparison is discussed later by using strain imaging results.

Fig. 6 (b) shows a comparison of Bragg-dip transmission spectra obtained from coarse grain (single crystal) samples as part of this experiment (HUNS) and in the previous work (J-PARC) [6]. In the HUNS case, the Bragg-dip depth is not deep, and Bragg-dip width is broad. This is due to both low wavelength-resolution (about 0.5% in the present experiment, while that of the J-PARC experiment was about 0.35%) and low beam angular divergence (the L/D ratio of this experiment was less than 60, but that of the J-PARC experiment was about 2400 [6]). Thus, it was found that the Bragg-dip analysis is easily affected by wavelength and angular resolutions in contrast to the Bragg-edge analysis. As a result, grain-orientation imaging was relatively difficult, as discussed below. Incidentally, the difference of Bragg-dip positions between this experiment and the previous work was caused by differences in sample alignment between the neutron beam direction and the normal direction of the sample plate. However, this effect is small for the grain-orientation imaging result (see Fig. 9).

Fig. 6. Comparison of (a) Bragg-edge profiles and (b) Bragg-dip profiles between the

decoupled thermal HUNS and J-PARC MLF BL10 “NOBORU”. The spectra were integrated over about 100 pixels in order to obtain better statistical data.

2.5. Quantitative imaging results

Figs. 7 and 8 show quantitative imaging results of average of d -spacing distribution, d_{110} , relating to macro-strain (Fig. 7), and the FWHM of the d -spacing distribution, w_{110} , relating to micro-strain (Fig. 8) for ferrite/martensite rods, obtained by this experiment (HUNS) and the previous work (J-PARC) [4]. Data from the present study were analyzed in the same way as that of the previous work [4]. Both Figs. 7 and 8 show the samples with quenched depths of 3 mm, 5 mm and 7 mm from left to right. Images in the upper half of each figure are HUNS results, those of the lower half are J-PARC results. Note that the pixel size of the imaging detector, the beam angular divergence, the accelerator power and the measurement time all differed between these experiments. In the J-PARC experiment, the pixel size was 55 μm owing to the MCP detector [14], the L/D ratio was 337, the accelerator power was 120 kW, and the measurement times were 22 hours for all samples and 7 hours for the open beam, respectively [4]. For this reason, the spatial resolution of the imaging results obtained at each facility is quite different.

It is remarkable that the values of d_{110} and w_{110} evaluated at both facilities are consistent. As shown in Fig. 7, quantitative imaging results at both HUNS and J-PARC indicate that d_{110} in the ferrite (center) region was 0.2026 nm (light-green color), and 0.2032 nm (red color) in the martensite (rim) region. In Fig. 8, w_{110} in the ferrite region is seen to be less than 0.001 nm (blue color), and 0.002 ~ 0.004 nm (green ~ red color) in the martensite (rim) region, both of which were correctly visualized by both the HUNS and the J-PARC experiments. In addition, in both the d_{110} and the w_{110} imaging, HUNS was able to visualize the radial dependence of quenched depth for each rod (3 mm, 5 mm and 7 mm). These results confirm that a pulsed CANS used with a decoupled moderator is effective for quantitative strain imaging.

Fig. 7. Comparison of quantitative imaging results of average of d -spacing (crystal lattice plane spacing) distribution, relating to macro-strain, performed at HUNS and J-PARC. The J-PARC results were reported in Ref. [4].

Fig. 8. Comparison of quantitative imaging results of FWHM of d -spacing (crystal lattice plane spacing) distribution, relating to micro-strain, performed at HUNS and J-PARC. The J-PARC results were reported in Ref. [4].

Fig. 9 shows the results of inverse pole figure (IPF) mapping to image grain orientation, using the Bragg-dip transmission method; the data shown were obtained by this experiment (HUNS) and the previous work (J-PARC) [6]. The grain-orientation image obtained at J-PARC was clearly visualized. Note that the beam angular divergence, the accelerator power and the measurement time were different between these experiments. In the J-PARC experiment, the L/D ratio was 2400, the accelerator power was 500 kW, and the measurement times were 14.5 hours for the sample beam and 7.2 hours for the open beam [6]. On the other hand, the results obtained at HUNS were relatively noisy (low statistics) due to the shallow Bragg-dips as discussed in Fig. 6 (b). In addition, since the bottom side of Fig. 9 (a) corresponds to the outside region of the supermirror guide-tube, the statistical error becomes larger than on the upper side. However, the grain orientations evaluated at HUNS were consistent with those evaluated at J-PARC, although the sample alignments were slightly different as discussed above. In any case, it was confirmed that grain-orientation imaging using the Bragg-dip transmission method

is also feasible at a pulsed CANS using a decoupled moderator, although the Bragg-dip analysis is more difficult than the Bragg-edge analysis due to low wavelength and angular resolutions.

Fig. 9. Comparison of quantitative results of grain-orientation imaging, using an inverse pole figure (IPF). Images were obtained at (a) HUNS and (b) J-PARC. The white dashed lines of the HUNS result correspond to the separation positions between the sample pieces. Fig. 9 (b) (the J-PARC result) is reproduced with permission of the International Union of Crystallography from Ref. [6].

3. Preliminary Monte-Carlo simulation study of a cold-neutron moderator with decoupled pre-moderators and poisoned pre-moderators

In the previous section, we report that the strain imaging using the Bragg-edge method and the grain-orientation imaging using the Bragg-dip method are feasible at a pulsed CANS using a decoupled thermal-neutron moderator. However, although a supermirror guide-tube was used, the neutron flux was still insufficient, suggesting the need for a cold-neutron moderator. As such a moderator, one can use a multiple-stage moderation system, like a coupled moderator consisting of a cold-neutron main-moderator combined with a thermal-neutron pre-moderator. This work investigated two possibilities for an improved system: a cold-neutron main-moderator with a thermal-neutron pre-moderator and a decoupler, and a cold-neutron main-moderator with a poisoned pre-moderator. The cold-neutron pulse characteristics of these new moderator systems were analyzed via a Monte-Carlo simulation, and the results compared with conventional coupled, decoupled and poisoned cold-neutron moderators.

3.1. Calculation method, calculation geometry, and evaluation method

The Monte-Carlo neutron transport simulation code used in this calculation was the Particle and Heavy Ion Transport Code System (PHITS) Version 2.820 [15]. We compared the neutron pulse shapes (neutron emission time distributions) of cold neutrons (5 meV) for five types of moderator.

The target material was lead ($7\text{ cm} \times 7\text{ cm} \times 7\text{ cm}$) as a photo-neutron source (black materials in Figs. 10 and 11). The reflector material was graphite (gray materials in Figs. 10 and 11), and the reflector thickness was 40 cm, which can provide neutrons of saturated intensity.

Fig. 10 shows the conventional moderator system designs for obtaining reference data. The main-moderator for all types was 20 K methane of 5 cm thickness and $10\text{ cm} \times 10\text{ cm}$ area. The coupled moderator has a polyethylene pre-moderator of 1.5 cm thickness at 300 K. The decoupled moderator used in the simulation has a cadmium decoupler of 1 mm thickness instead of a pre-moderator. The decoupled-poisoned moderator contains a cadmium plate of 1 mm thickness at the half thickness (2.5 cm) position in the moderator. These moderator geometries were adopted for HUNS to maximize the total neutron flux, and were not optimized in terms of both brightness for high L/D experiments and wavelength resolution.

Fig. 10. Design of moderators used in simulation study. (a) A coupled cold-neutron moderator; (b) a decoupled cold-neutron moderator; (c) a decoupled-poisoned cold-neutron moderator. The dimensions of the main-moderator (20 K methane) are 5 cm in thickness, 10 cm in height, and 10 cm in width.

Fig. 11 (a) shows a new type of moderator; a cold-neutron moderator with decoupled pre-moderators. This geometry corresponds to a coupled moderator with a decoupler. This

3.2. Results and discussion

Fig. 12 shows plots of cold neutron fluxes using each of the moderator systems; Fig. 12 (a) is plotted using a linear scale to evaluate the peak intensity and the pulse width, and Fig. 12 (b) employs a semi-log scale to evaluate the pulse decay. Table 1 presents a comparison of the moderator systems in terms of the peak intensity, pulse width (FWHM), and pulse decay (1/10 pulse width). The two types of moderator proposed in this work have the highest peak intensity (higher than a conventional coupled moderator). On the other hand, these new moderator systems have narrow pulse widths, about the same as those achieved by the decoupled moderator (60 μ s) and the poisoned moderator (40 μ s) (see Table 1). In addition, the decay time of these new moderator systems match that of the poisoned moderator (about 120 μ s, see Fig. 12 (b) and Table 1). The reason that the poisoned pre-moderator system has a narrower pulse width than the decoupled pre-moderator system is the thickness of the inner pre-moderator. In other words, as mentioned above, the backside pre-moderator can be made thinner (from 2 cm to 1 cm) owing to the presence of the poison layer. The reason that the poisoned pre-moderator system has the same peak intensity as the decoupled pre-moderator system is the existence of the outer pre-moderators.

This study suggests the possibility of new moderator systems which simultaneously use both a pre-moderator and a decoupler (or poison), and it is feasible that both higher peak intensity and sharper neutron pulses can be simultaneously achieved in terms of high brightness (high L/D experiment). This can help to improve the “shallow dip depth” problem in a Bragg-dip neutron transmission imaging experiment at a pulsed CANS.

Fig. 12. Cold neutron emission time distributions for each moderator system, expressed on (a) a linear scale and (b) a semi-log scale.

Table 1. Comparison of peak intensity, pulse width (FWHM), and decay time (1/10 pulse

width) of cold-neutron pulses for each moderator system.

Moderator type	Peak intensity (n/cm ² /MeV/source)	Pulse width (FWHM) (μs)	1/10 pulse width (μs)
Coupled moderator	225	80	400
Decoupled moderator	175	60	200
Poisoned moderator	175	40	125
Decoupled pre-moderator system	300	55	130
Poisoned pre-moderator system	300	40	110

4. Conclusion

By using a supermirror guide-tube coupled to a decoupled thermal-neutron moderator (polyethylene at ambient temperature), we achieved strain imaging using the Bragg-edge transmission method and grain-orientation imaging using the Bragg-dip transmission method, under the condition of 0.5% wavelength resolution at a compact-accelerator driven pulsed-neutron source. Although there is room for improvement in neutron flux and neutron beam angular divergence (in particular, for the Bragg-dip transmission method), measured strain values and grain orientations agreed well with the results of the J-PARC experiments.

Monte-Carlo simulations were performed to evaluate the performance of a thin and low-height cold-neutron moderator (20 K methane) with the new components, a decoupled pre-moderator and a poisoned pre-moderator with large solid-angle coverage for fast neutrons. The results suggest that these components offer higher peak intensity than a coupled moderator (about 1.3 times higher), the same pulse width as decoupled/poisoned moderators (about 60 or 40 μs), and the same decay time as a poisoned moderator (about 120 μs). Note that these values were attained only over a certain region (1 cm × 1 cm) of highest neutron brightness. In other words, these moderator modifications are suitable for high L/D experiments with low beam angular divergence using a pinhole collimator. Further detailed investigations are necessary to more fully characterize these new types of moderator systems.

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