In this letter we show a spectral and x-ray intensity analysis that unequivocally proves a spectrally resolved plasmon reported in *Fletcher et al* [1]. The analysis takes the f-sum rules into account [2, 3], is consistent with the independently and simultaneously measured wave-number resolved x-ray scattering signal, and compares well with advanced elastic feature modeling including density functional theory for molecular dynamics (DFTMD) [1] as well as pseudo-atom molecular dynamics [4].

Figure 1a is plotted to demonstrate the proper comparison of the backward spectrally resolved scattering signal with the forward spectrally resolved scattering signal reported in *Fletcher et al*. A fluorescence calibration will show the backscatter spectrometer is out of focus, and depth broadened via crystal thickness [5], by 2x the forward spectrometer [*see supplemental*]. An analysis that does not account for this would be misleading and incorrect. Applying a simple ray tracing adjustment, we find that there are no observable low-energy features in the backscatter spectrometer that are at the same spectral location as that of the plasmon in the forward spectrometer. Additionally, the elastically scattered intensity matches the theoretical prediction for the back-to-forward ratio [1, 4] for the shock conditions inferred in the experiment.

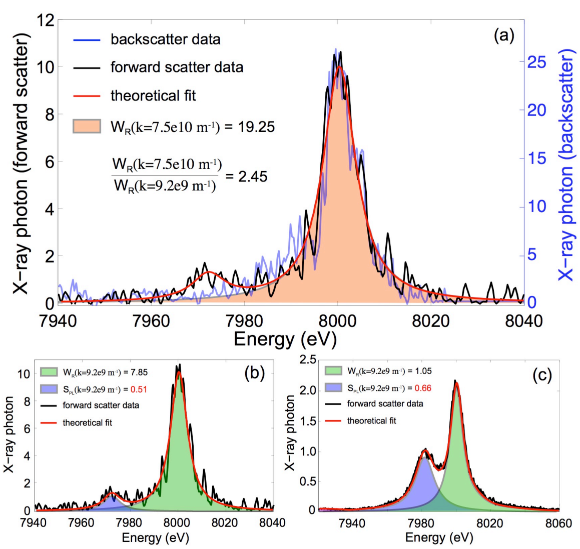

Figure 1 – a.) Backscatter signal (blue) is plotted with the forward spectrum (black), and the theoretical fit (red) to the data. Right axis (blue) is the x-ray photons measured for a single shot in the backscatter data. Left axis (black) is the x-ray photons measured for a single shot in the forward scatter data and the expected signal of the backscatter data as observed on the forward spectrometer based on the 2.45x decrease in the integrated elastically scattered signal $W_R(k)$ (shaded orange); In b.) we show a single shot forward spectrum (black) together with the best fit source function signal (shaded green), the plasmon signal (shaded blue), and the theoretical fit to the forward spectrum (red line). The fit is calculated for ρ=2.32 ρ_0, and T_e=1.75 eV. In c.) we show a multi-shot averaged forward scattering spectrum (black), the best fit source function signal (shaded green), the plasmon signal (shaded blue), and the theoretical fit to the forward spectrum (red line) where best fit is achieved at conditions of $\rho=\rho_0$, (x-ray only). The spectra in a), b) and c) clearly shows down-shifted plasmon signals in the forward spectrometer with no plasmon in the backscatter spectrometer as expected; a similar x-ray scattering spectrum, for an identical experimental configuration, has been previously reported [6].

The spectra in *Fletcher et al* were fit with theoretical models that correctly account for the f-sum rules. As observed in Figure 1b and 1c, the overall inelastic shift leads to a decrease in intensity by 29% from the x-ray only plasmon. Such a signal decrease is predicted for the drop in the plasmon intensity when going from x-ray only ($\rho=\rho_o$) to laser shock compressed conditions (T_e=1.75 eV, ρ=2.32 ρ_o), as reflected in the theoretical fits for figure 1b and 1c.

The majority of the compression data collected in this experiment were done under shock conditions (single sided laser irradiation) that results in compression states that are not high enough to overcome the spectral bandwidth of the SASE x-ray laser beam and crystal spectrometers for the inferred temperature states. The only way to unequivocally resolve a plasmon signal for the material conditions, and experimental configuration, specified in the manuscript [1] is with 1.) the seeded x-ray laser, and 2.) with compression states that shift the plasmon signal far enough away from the elastic scattering feature (i.e. $\rho/\rho_0 > 1.5$). Within these straight-forward criteria, the necessary photometrics (i.e. x-ray laser photons per pulse) have to also be met simultaneously. The histogram plotted in Figure 2, demonstrates the measured x-ray laser pulse energy for each shot that meets these two criteria. The figure clearly shows that single shot plasmons can only be observed for total energies that are above 1.6 mJ. The data set highlighted in green represents shots where a plasmon was observed in the spectrally-resolved forward scattering spectrometer and no backscatter features at the same scattered energy were present. In our case, we have agreement for two plasmons that are both independently verified by the structure factor (as measured with wide angle x-ray scattering) for two different material conditions. The sampling statistics, and photometric calculations are consistent with the theoretical framework for such a study. The dark green indicates a plasmon signal that was not included in the final analysis because the S(k) data indicated a significant gradient in the temperature and compression states resulting in a data set that was not well constrained and therefore appropriately removed.

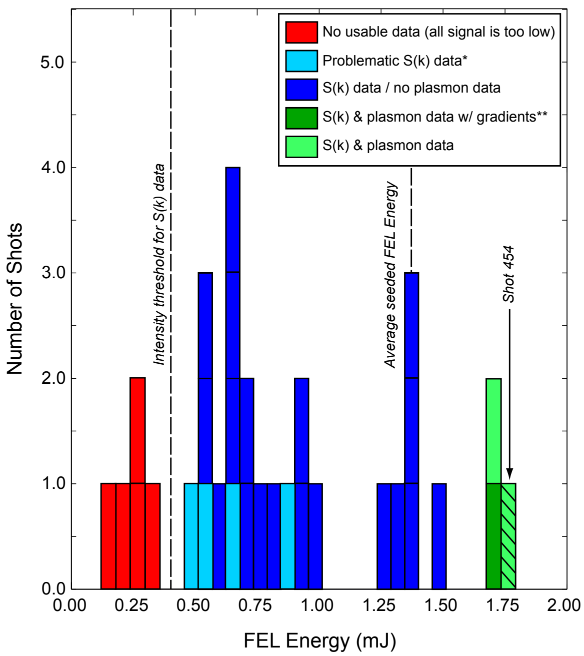

Figure 2 –Histogram of seeded x-ray mode with material states above 1.5x compression demonstrating the criteria used for single shot spectral analysis. The data in red corresponds to shots where the x-ray laser intensity was too low to observe any usable data for S(k) and spectral analysis. The data in blue corresponds to shots where S(k) data is clearly observed; (dark blue) corresponds to S(k) data and XRTS-elastic data included in the analysis, (light blue*) is data that shows multiple, unconstrained, and/or inhomogeneous material states from wide angle scattering. The shots in green show both S(k) data and plasmon data; (dark green**) is data where significant gradients are observed in the wide angle scattering, thus resulting in unconstrained spectrally resolved signals; (light green) are data that show simultaneous angularly and spectrally resolved plasmon signals that were reported in *Fletcher et. al*. Shot 454 has the highest x-ray laser intensity of all the data collected under this configuration. The average x-ray laser energy was determined by all seeded mode x-ray energies measured throughout the campaign.

The stability of the x-ray laser intensity (using seeding mode), was a significant contributing factor to the spectral data utilized in *Fletcher et al*. As it can be observed in Figure 2, all x-ray laser energies below 1.6 mJ do not demonstrate an observable single shot plasmon signal (blue data). The wide angle scattering data, however, is dominated by elastic scattering from bound and weakly bound electrons that have a cross section that is 15-200 times larger (depending on the 2θ scattering angle) than the scattering contribution from collective free electron oscillations (plasmons). This results in data where the wide-angle x-ray scattering data can be used in the analysis, but the spectrally-resolved plasmon signal has insufficient photometrics (dark blue data in Figure 1). There are also circumstances (red data in Figure 1) where the x-ray laser energy per pulse is too low to observe usable signals for a proper analysis. The availability of plasmon data is clearly labeled in Figure 5 of *Fletcher et al*.

While a large number of shots maybe executed at "identical" design conditions of the experiment, many of the shots may result in different material states on a shot-to-shot basis due to fluctuations in the drive laser intensity, shock and x-ray probe timing, or intended variability in the target design. All of these circumstances can be determined from the large set of available diagnostics, from supplemental measurements in our experimental configuration, and have been properly accounted for in the data analysis. There are also shots were the laser-generated shock waves are mis-aligned, or the drive laser misfires, thereby producing multiple and/or unconstrained material states. Such inhomogeneous conditions, and density/temperature gradients, can be identified within the suite of diagnostics fielded in this study (e.g., wide angle scattering or Compton scattering), thus requiring selection/removal and analysis of appropriate data points. These data are labeled in light blue in Figure 2.

Based on the analysis presented by *Gamboa*, examining the calculated elastically scattered signal levels would indicate a state of matter that has a temperature exceeding 10 eV [1, 4]. *Gamboa* fails to disclose that his interpretation of all the spectra results in a violation of basic shock physics and plasma theory [2, 7]. The data, as interpreted by *Gamboa*, would require much higher densities and temperature states than can be reasonably expected and would not be consistent with the angularly resolved S(k) diagnostic. However, the plasmon signal measured in *Fletcher et al* is supporting independent S(k) diagnostic that directly measures the structure factor via diffraction. This S(k) data agrees with the plasmon signal as well as the elastic scattered amplitudes that are presented in the manuscript. Furthermore, the integrated elastically scattered signals present in the spectral analysis (along with simultaneous S(k) data), have been independently verified using DFTMD [1], and pseudoatom molecular dynamics [4].

In December 2018, a successful experiment was concluded at the Linac Coherent Light Source (LCLS) (experiment LU52) on laser shock-compressed Cu where a measured, well-pronounced, plasmon feature whose frequency is sensitive to compression was observed. This observation provides strong evidence for the utility of plasmon scattering. In addition, the range of densities and pressures found in LU52 supports the findings on compressed aluminum. Thus, we are absolutely confident that the aluminum data are correct and that the conclusions of *Fletcher et al* are valid.

References

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Supplemental information

The procedure in *Fletcher et al*, uses copper K-α fluorescence, along with single hit analysis, to compare the signal levels of the forward inelastic plasmon signal, and how it compares to any elastically scattered features present in the backscattering spectrometer. Analysis of the fluorescence signal reveals the correct relative signal levels recorded on the two spectrometers as well as the ability to observe and characterize the alignment and depth broadening of both spectrometers, where the backscatter crystal spectrometer has a spectral bandwidth that is two times larger than the forward spectrometer (Figure 1S).

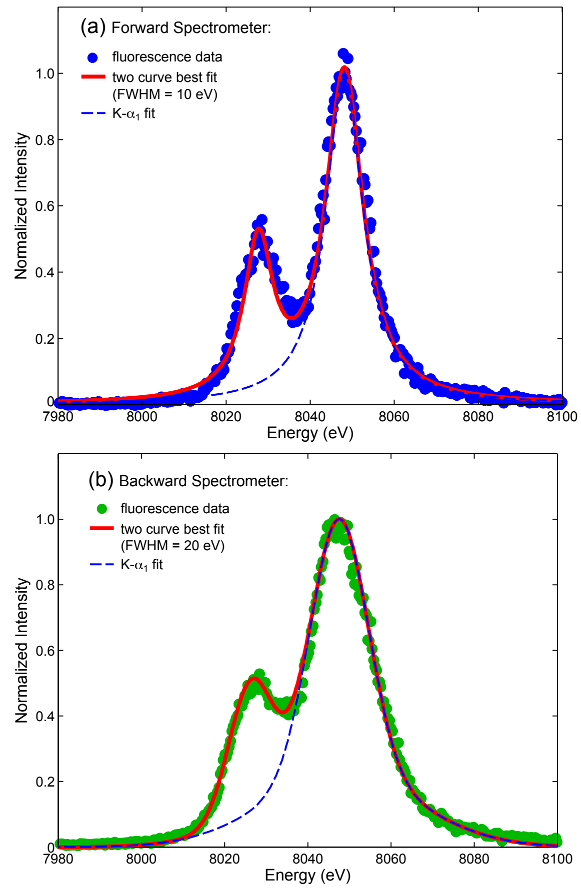

Figure 1S – a.) Normalized Cu K-α fluorescence measured on the forward spectrometer (blue dots), and the best fit to the K-α doublet (red). The best fit to the K-α_1 (blue dashed line) shows a spectral resolution of 10 eV (FWHM); b.) Normalized Cu K-α fluorescence measured on the backward spectrometer (green dots), and the best fit to the K-α doublet (red). The best fit to the K-α_1 (blue dashed line) shows a spectral resolution of 20 eV (FWHM). Such a discrepancy in the spectral resolution of the back spectrometer is caused by slight misalignment and x-ray signal broadening caused by different crystal thicknesses; where (a) is 40 μm thick and (b) is 100 μm thick. The source function in the backward spectrometer is two times broader spectrally than it is measured in the forward spectrometer.