The manuscript presents FDTD simulations of the polarization dependent emission localization from quantum dots near an L-shaped nanostructure. The simulations indicate that distant dependent coupling between the QD and nanostructure affect the apparent localization of the QD in a manner consistent with the polarization and distance from each branch of the nanostructure. The dimensions on which these effects are expected to manifest require measurements that can provide spatial and intensity information on length scales of less than 100 nm. It appears unfortunate that experiments to validate the simulations were unsuccessful, as stated in the manuscript on p. 16, lines 34-40 and noted at other places in the text. Without experimental validation, it is difficult to assess if the simulations provide a meaningful model or not. The following are issues and suggestions that would improve the clarity of the work.

1. There are results presented that are unclear if they arise from simulation or experiment. Figure 1 C shows 4 spectra that are attributed to something. These appear to be simulations; however, the caption is unclear. If these are experiments, does the spectral response provide another angle to assess to position of the QD relative to the nanostructure? The nature of the data needs to be clarified at a minimum.

2. Figure 3 appears to be experimental data; however, what it is conveying is not clear. Can this data be plotted in a way to better illustrate how the change in emission can be correlated to the position of the nanostructure? Perhaps plotting the centroid of each polarization angle on the same set of axis? Alternatively, is there information in the shape of the observed psf that may be informative? The figure as presented does not aid understanding of the effect.

3. How was the ground truth position of the nanoparticle determined in Figure 4b? Is this an unique instance where the correlated SEM experiment worked? The simulation and experiment are under different conditions and generate different polarization dependent intensities. There is a qualitative
agreement, but would simulating the exact experimental conditions provide better agreement? This should be addressed in revision.

4. The result in Figure 6b is unclear. This appears to plot how the polarization of antenna affects the emission intensity of the QD. The caption suggests this is a comparison between the simulated polarization dependent emission and the plasmon resonance spectrum. What is plotted is unclear, and may be a key element in understanding the observed phenomena.

5. Removing the dip-pen nanolithography is suggested. Figure S1 is supposed to illustrate improved placement of the QD; however, the figure shows the QD is not in the critical part of the L-shaped nanostructure. A future manuscript might be appropriate when the DPN is properly optimized.

Author’s Response to Peer Review Comments:

We would like to thank the reviewer and the editor for their comments and effort in making this manuscript better structured and clearer both scientifically and non-scientifically. Based on the comments, we separated the main manuscript and the supporting information (PlasmonicManuscriptUpdated.docx and PlasmonicManuscriptSI.docx), added in the TOC graph, and made both scientific non-scientific changes to the manuscript. The manuscript with markups (PlasmonicManuscriptMarkup.docx) is also attached to help the editor and reviewer follow the locations of revisions. Please see our response to the reviewer’s comments in PlasmonicReviewResponse.docx.
1. There are results presented that are unclear if they arise from simulation or experiment. Figure 1 C shows 4 spectra that are attributed to something. These appear to be simulations; however, the caption is unclear. If these are experiments, does the spectral response provide another angle to assess to position of the QD relative to the nanostructure? The nature of the data needs to be clarified at a minimum.

We thank the reviewer for the kind suggestion. Figure 1c represents the calculated scattering cross-sections of nanoantennas. We edited the caption of figure 1, stating in both the figure caption and main text that the results are from FDTD simulations.

2. Figure 3 appears to be experimental data; however, what it is conveying is not clear. Can this data be plotted in a way to better illustrate how the change in emission can be correlated to the position of the nanostructure? Perhaps plotting the centroid of each polarization angle on the same set of axis? Alternatively, is there information in the shape of the observed psf that may be informative? The figure as presented does not aid understanding of the effect.

Figure 3c is a set of fluorescence images captured at different emission polarizations for the same QD (red box in Figure 3b). The title of each image indicates the emission polarization angle and mislocalization of the emitter compared to the sum image. If we plot the change of fitted emitter center as a function of emission polarization, we will get a similar plot as Figure 4. Figure 3a and 3b indicate that there is QD next to the nanoantenna. However, we were not able to find the true location of QD due to SEM failure. We update the caption of Figure 3 based on the reviewer’s comment.

3. How was the ground truth position of the nanoparticle determined in Figure 4b? Is this an unique instance where the correlated SEM experiment worked? The simulation and experiment are under different conditions and generate different polarization dependent intensities. There is a qualitative agreement, but would simulating the exact experimental conditions provide better agreement? This should be addressed in revision.

We thank the reviewer for pointing out the part that needs clarification. The experimental emitter position labeled in Figure 4 is calculated by fitting the sum image of images captured at all polarizer and the analyzer polarizations. The emitter position input for the simulation is from the SEM image of the same nanoantenna/QD system. This position difference shows the mislocalization in fluorescence measurement. The matching pattern between simulation and experiment plots shows the feasibility of using simulation to study the complex interaction between nanoantenna and QD. However, we agree that this statement
needs further support from more measurements (which is discussed in the manuscript). We update the caption of Figure 4 based on the reviewer's comment.

4. The result in Figure 6b is unclear. This appears to plot how the polarization of antenna affects the emission intensity of the QD. The caption suggests this is a comparison between the simulated polarization dependent emission and the plasmon resonance spectrum. What is plotted is unclear, and may be a key element in understanding the observed phenomena. Figure 6 are calculated results that demonstrate how apparent polarization angle changes for three different emitter dipole orientations and how it is determined by the nanoantenna scattering. The scattering from the plasmonic interface is believed to be the key component that contributes to the rotation of QD emission polarization because two spectra in Figure 6b have matching patterns. Thanks to the reviewer's comment, this information is emphasized in the figure caption.

5. Removing the dip-pen nanolithography is suggested. Figure S1 is supposed to illustrate improved placement of the QD; however, the figure shows the QD is not in the critical part of the L-shaped nanostructure. A future manuscript might be appropriate when the DPN is properly optimized. We thank the reviewer for the kind suggestion. We would like to keep figure S1 because it points to improved success rates and more meaningful statistical results for this experiment. We fully admit that we did not achieve the desired QD position with DPN (this non-precise positioning is now fully disclosed and emphasized in S1 caption), but want to motivate future studies by us and others to exploit this route. Thus, a detailed discussion on how to improve the success rate and throughput has also been presented in the manuscript.
Name: Peer Review Information for "Super-Resolution Imaging of Plasmonic Near-Fields: Overcoming Emitter Mislocalizations"

Second Round of Reviewer Comments

Reviewer: 1

Comments to the Author
1. What is the major advance reported in the paper?

The revised manuscript improves the clarity of the figures and what is being reported. The manuscript claims that polarization dependent changes in the localization occur due to interactions between the arms of the L-shaped nanostructure and the position of a quantum dot (QD) within the L. The proposed model would increase understanding and the utility of localization microscopy. However, there are significant questions regarding the reported data and the ability to use this effect to understand the position of the QD which should be addressed prior to publication. For reasons listed below, it is not clear the effect is observed experimentally or that the effect suffers from artifacts that should be clarified.

2. What is the immediate significance of this advance?

Localization microscopy is seeing increasing use for investigating spatial interactions using far-field collected radiation to explore nanoscale phenomena. This report investigates interactions between nanostructures that alter the observed localization vs the actual localization. The proposed theory, if it can be validated may be useful.

3. Technical suggestions

In Figure 3, if you plot out the coordinates of the polarization emission, you get the following plot:

The displacement (effective change in localization) does not appear to correlate with the polarization angle. Polarization angles that have the same direction do not agree, and some results seem anomalous. Can this be explained? How has beam displacement as the polarizer is rotated accounted for?

Note: The table generated from the data in Figure 3 is included to assist the authors.
The manuscript notes multiple experimental difficulties that will need to be accounted for in future work on p. 29, lines 3-7 (noting differences in the simulated and observed spectra in Figure 6), p. 30, line 24 (further examining the change in \( \theta_{\text{app}} \) and scattering efficiency in Table 1), and p. 24 line 4 (regarding the inability to use SEM to find the real position of the emitter). It is not clear that the imaging experiments have proper controls.

In Figure 4, What does Simulation (107 nm, 37 nm) and Experiment (81 nm, 28 nm) over the plots represent? The text (p. 19 lines 32-35) indicates “The emitter position input for the simulation is from the SEM image of the same nanoantenna/QD system.” However, on p. 24 the manuscript says the SEM experiment has not worked. Additionally, the notation suggests the position, which may be incorrect, but why not simulate the same position as the experiment?

The revised manuscript now mentions improved QD placement by dip pen nanolithography (DPN) 3 times, on p. 11 lines 3-7, p. 24 line 21-25, and p. 36 lines 7-14. On p. 36, the manuscript indicates a higher success rate. However, figure S1 (below) only shows a QD completely outside the L. The statements are not supported.

The DPN seems to be something the authors would like to work, perhaps claim, but cannot substantiate.

Author’s Response to Peer Review Comments:
We would like to thank the reviewer and editor for their feedback and valuable comments. We made modifications to the manuscript to address the reviewer’s comments and concerns. We are also attaching a rebuttal letter to further explain our modifications and what we worked on to improve our presentation.
We thank the reviewers for all the kind comments and constructive advice. Please see below the reviewer’s comments in black, and our responses in light purple.

1. The displacement (effective change in localization) does not appear to correlate with the polarization angle. Polarization angles that have the same direction do not agree, and some results seem anomalous. Can this be explained? How has beam displacement as the polarizer is rotated accounted for?

   We thank the reviewer for the suggestion. For the QD in the red square, we did see that the displacement has weak correlation with the polarization angle. However, we do have plots that shows close correlation (updated Figure 3c and Figure 4b). The reason that this plot is not in the original manuscript is that we did not have matching Figure 3a and 3b for the plot. We have revised the figure caption based on the updated plot.

2. The manuscript notes multiple experimental difficulties that will need to be accounted for in future work on p. 29, lines 3-7 (noting differences in the simulated and observed spectra in Figure 6), p. 30, line 24 (further examining the change in $\theta_{app}$ and scattering efficiency in Table 1), and p. 24 line 4 (regarding the inability to use SEM to find the real position of the emitter). It is not clear that the imaging experiments have proper controls.

   We thank the reviewer for the concerns. Controls are used to make sure the true QD center displacement is measured. The displacement of QDs next to the nanoantenna are corrected with positions of single QD without nanoantenna nearby. This control is served as fiducial markers for drift correction, and since the dipole orientation of QD alone is degenerate, QD localization is independent of the excitation and emission polarizations. Meanwhile, the final displacement plot (Figure 4) is the average over four full circles of the polarizer rotation.

3. In Figure 4, What does Simulation (107 nm, 37 nm) and Experiment (81 nm, 28 nm) over the plots represent? The text (p. 19 lines 32-35) indicates “The emitter position input for the simulation is from the SEM image of the same nanoantenna/QD system.” However, on p. 24 the manuscript says the SEM experiment has not worked. Additionally, the notation suggests the position, which may be incorrect, but why not simulate the same position as the experiment?

   The experimental emitter position (81 nm, 28 nm) is the fitted center of the sum image of all polarizer and the analyzer polarizations. The emitter center for simulation (107 nm 37 nm) is from the SEM image of the same nanoantenna/QD system. This information is updated in both the figure caption and the main context.

   We thank the reviewer for pointing out the inconsistency in the context. Figure 4b is from the only success iteration that closes the loop of sample fabrication, QD deposition and SEM making out singe QD. We were not able to acquire enough experimental results so claimed that ‘the SEM experiment has not worked’. We have updated the context on p. 24 to clarify this statement.

   We believe that SEM provides us the true location of the emitter (107, 37 nm), so we use it as the input for the simulation (instead of the fitted center from the sum image
This difference in QD centers between SEM and optical measurement in fact validates the mislocalization phenomenon and supports the importance of our study.

4. The revised manuscript now mentions improved QD placement by dip pen nanolithography (DPN) 3 times, on p. 11 lines 3-7, p. 24 line 21-25, and p. 36 lines 7-14. On p. 36, the manuscript indicates a higher success rate. However, figure S1 (below) only shows a QD completely outside the L. The statements are not supported.

We thank the reviewer for the kind suggestion. We fully admit that we did not achieve the desired QD position with DPN and have disclosed and emphasized this non-precise positioning in the caption of Figure S1. We removed the claim of ‘higher success rate’ and moved the DPN method description to the supporting information. We do believe that DPN provides a more precise QD deposition since it replaces random QD positioning (our original method in the manuscript) with a controlled strategy. A detailed discussion on how to potentially improve the success rate and throughput of DPN has also been presented in the SI. We would like to keep the DPN part to motivate future studies by us and others to exploit this route.
Comments to the Author

The insistence on including the dip pen nanolithography (DPN) work is confusing. The data in Figure S1 shows the approach did not work. The authors admit in the response letter it has not worked, yet unsubstantiated statements remain in the paper that the method is better. The statement at the top of p. 11, indicating DPN provides higher precision needs to be removed.

The statement on p. 18 lines 34-41, should be revised to indicate the SEM validation is not realized in most experiments, since this was only successfully done 1 time.

I believe the modeling likely captures the experiment; however, it is difficult to make any conclusions on a single validated measurement.

Author's Response to Peer Review Comments:

We thank the reviewers for all the kind comments and constructive advice. We have made edits to the manuscript based on the comments. Please see our response in the attached file.
We thank the reviewers for all the kind comments and constructive advice. Please see below the reviewer’s comments in black, and our responses in light purple.

1. The insistence on including the dip pen nanolithography (DPN) work is confusing. The data in Figure S1 shows the approach did not work. The authors admit in the response letter it has not worked, yet unsubstantiated statements remain in the paper that the method is better. The statement at the top of p. 11, indicating DPN provides higher precision needs to be removed.

   We thank the reviewer for the suggestion. We fully admit that we did not achieve the desired QD position with DPN and have disclosed and emphasized this non-precise positioning in the caption of Figure S1. We removed all the claim of ‘higher success rate’ in both main text and supporting materials and refer to DPN as an alternative sample preparation method that we tested. A detailed discussion on how to potentially improve the success rate and throughput of DPN has also been presented in the SI. We admit that both 2-step photolithography and DPN need further development and improvements to reach ~10nm accuracy of positioning QDs with respect to the antenna. Both approaches are now presented as alternatives that need improvements.

2. The statement on p. 18 lines 34-41, should be revised to indicate the SEM validation is not realized in most experiments, since this was only successfully done 1 time.

   We thank the reviewer for the suggestion. We have updated the statement and disclose the low success rate in the context.

3. I believe the modeling likely captures the experiment; however, it is difficult to make any conclusions on a single validated measurement.

   Yes, we fully agree with the reviewer’s comment. We admit that our result might not be statistically significant, but it shows the correspondence between modeling and experiment and shows potential for further investigations. We realize that our study has limitations due to the low success rate, and provide ideas in the manuscript on what could be done to improve the experiments. We hope that our study could trigger more interests and provide insight for future studies that will establish good statistical confidence and better correlation between theory and experiments.