**Lists of the revised part**

Corrections are colored in red in the “marked up manuscript.” The color is cleared in the “(revised) article file.”

I. Introduction

* We have replaced the terms “irradiative” by “non-radiative” throughout the paper.
* We have added a sentence to present that non-radiative damping of SPP is termed as the Ohmic loss in the fourth paragraph.

II. Device Design and Fabrication

* We have corrected the misspelling of “height” at the end of the sixth paragraph and the misspelling of “evaporation” at the end of the seventh paragraph.

IV. Results

* We have corrected the misspelling of “below” at the end of third paragraph.
* We have added the comments on the difference between our observations on gold gratings and observations by Cleary et al. on silver gratings, at the end of the section.

V. Discussions

* A paragraph has been added to explicitly show how the dielectric constant depends on the Drude parameters, as the second paragraph.
* The expression for the wavenumber of SPP has been corrected in the third paragraph: (/2) has been corrected to (2)/ and tilde has been added on top of , at the third paragraph.
* A paragraph has been added to explicitly show how LSPP depends on the dielectric constant and to show  > , as the fourth paragraph.
* Several sentences have been modified to introduce more clearly the findings in [Trollmann et al., JPCC 2014], in the fifth paragraph.
* Several sentences have been added to present more precisely the interpretation of our experimental results, in the sixth paragraph.
* A paragraph including Eqn. (5) has been added to show how LSPP depends on the electron scattering rate , and to explain the underlying physics, as the seventh paragraph.
* In the eighth paragraph, we have replaced “scattering hotspots” by “electromagnetic hotspots,” “SPP scattering” by “SPP attenuation,” and “multiple SPP scattering” by “multiple SPP reflection within the hotspots.”

Other changes

* The order of authors have been changed.
* An acknowledgement to K. Edagawa has been added.
* Two references have been added: Peale et al, JOSAB 2008 and Raether (Springer-Berlin, 1998)
* We have corrected and completed the information Ref. 5 and Ref.22.

**Reply to Reviewer 1**

**[Reviewer 1’s Comments #1]**

*This paper measures propagation length of infrared surface plasmon polaritons on gold. It shows that the propagating length significantly increases with annealing. The increase is correlated with the morphology becoming less granualar. While the result is not surprising, I’m not aware of a comparable experimental demonstration, so the work is useful. The work should be published, but I strongly urge including some additional transport data to support the interpretation..*

**[Our responses]**

We thank Reviewer 1 for careful reading and his/her valuable comments. Our responses to the comments and the corresponding changes are described below.

**[Reviewer 1’s Comments #2]**

*The authors attribute the increase in propagation length to a decrease in Ohmic losses. The theory presented (Eq. 2) shows that propagation length is determined from the complex permittivity, but there is no connection between the complex permittivity and “Ohmic losses”. Thus, while the explanation sounds reasonable, the connection to film resistivity is hand‐waving at best.*

**[Our responses]**

Scattering of free electrons (by electrons, phonons, defects, impurities, crystallite grain boundaries, etc.) causes the relaxation of collective oscillation of free electrons and the resultant non-radiative damping of SPPs. Energy loss of SPP due to the non-radiative damping” ends up with the Joule heat, and therefore it is usually called as the Ohmic loss. Therefore the measure of the Ohmic loss is the propagation length of SPP determined by the non-radiative damping. In this regard, the connection between the complex permittivity and the Ohmic loss has been described by Eqn.(3) (it was Eqn.(2) in the original submission).

In order to show this connection more explicitly, we have added Eqn.(4) in Section V.

Furthermore, we have added Eqn.(2) and (5) in Section V to show how LSPP depends on the electron scattering rate .

**[Reviewer 1’s Comments #3]**

*At ~5x longer wavelength, gold enters the regime of the quasi‐static approximation, where the imaginary part of the permittivity is proportional to σ/ω (e.g. see Soref et al Ref. 8). In that region, losses increase with increasing conductivity. In other words, “Ohmic” effects can cause more or less loss depending on the wavelength regime. Where the cross over occurs depends on the morphology of Au, e.g. for highly porous gold black the quasi‐static approximation down to at least 20 micron wavelength [JAP 118, 154307 (2015)].*

**[Our responses]**

The Joule heat induced by an oscillating electric field of frequency  is proportional to ’, the real part of the electric conductivity  = ’ + *i*”. Therefore the local Joule-heating efficiency inside metal is also proportional to ’ at any frequency (by the way, ’ is proportional to ”). Here ’has its frequency dependence characteristic to the Drude model, and the behavior of ’ upon decrease in the electron scattering rate  depends on whether or not the operating frequency  is higher than  (which Reviewer 1 calls as “the cross over”).

Here we added a sentence to explain that  >  is fulfilled in our experiments, referring to the data of the electron scattering rate for evaporated gold films with varied morphology [Trollmann et al. JPCC 2014].

**[Our related comments]**

We would like to give some comments about what determines the amount of the Ohmic loss. Although the local Joule heating efficiency is proportional to ’ as described above, the total Ohmic loss due to SPP depends not only on ’ but also on “the fraction of electromagnetic energy contained inside gold.” If large portion of the electromagnetic energy of SPP is contained in air (not inside metal), the total Ohmic loss becomes small, even if the local Joule-heating efficiency is large. This is why SPP’s propagate longer at lower frequencies of mid-infrared and terahertz range than at visible and near-infrared range.

In our experiments, thermal annealing increased the grain size and therefore decreased the electron scattering rate  (the correlation between grain size and  has been established in [Trollmann et al. JPCC 2014]. Equation (5) shows that LSPP should increase with decreasing . Two kinds of the underlying physics are the suppression of the local Joule heating (or suppression in ’) and the increase in the energy portion contained outside metal.

Here we have added a paragraph as the seventh paragraph in Section V, to explain the reason why *L*SPP increases with .

**[Reviewer 1’s Comments #4]**

*The authors provide no data from any measurement of resistivity as a function of annealing. That would be an easy result to add, and it would confirm the supposed correlation between SPP propagation length and conductivity. Otherwise, an alternate explanation could be that losses are radiative due to surface roughness.*

As described above, LSPP is determined not only by the real part of the conductivity s’ but also by the fraction of electromagnetic energy contained inside gold. Furthermore, the grain size is a more reliable parameter to describe the morphology than annealing temperature (annealing at the same temperature may result in different grain size, depending on the original morphology). Therefore it is more important to correlate the Drude parameters (especially ) with grain size, rather than to correlate resistivity with annealing temperature.

Here we have added several sentences to introduce the experimental data of the electron scattering rate in correlation to grain size from [Trollmann et al. JPCC 2014] in the fifth paragraph in Section V.

Some minor considerations:

[Reviewer 1’s Comments #5]Second paragraph after Fig.1: Misspelling of “height”. “The waveguides and the gratings were fabricated to have a common *height* of 0.8 μm from a gold base layer.”

[Our response]Following Reviewer 1’s comment, we have corrected the misspelling at the end of the 6th paragraph of section II.

[Reviewer 1’s Comments #6] Third paragraph after Fig. 1: “Evaporation”. “During the *evaporation* process, the substrate was not heated.”

[Our response]Following the comment, we have corrected the misspelling at the end of the seventh paragraph of section II.

[Reviewer 1’s Comments #7] Complete and correct citation for Ref. 5 is DOI: http://dx.doi.org/10.1557/PROC-1133-AA10-03. The MRS Proc volume number is 1133. The rest of the identifier for this online-only publication is AA10-03.

[Our response] Following the comment, we have completed and corrected the citation for Ref. 5.

[Reviewer 1’s Comments #8] Complete and correct citation for Ref. 21 is Long-wave infrared surface plasmon grating coupler , J. W. Cleary, G. Medhi, R. E. Peale, and W. R. Buchwald, Appl. Optics 49, 3102-3110 (2010). Specifically, the authors have left out Dr. Medhi’s last name.

[Our response] Following the comment, we have completed and corrected the corresponding citation. In the revised manuscript it is numbered as #23.

[Reviewer 1’s Comments #9] A different study of SPP propagation in a different wavelength regime, which the authors may find relevant: Propagation of high‐frequency surface plasmons on gold , R. E. Peale, O. Lopatiuk, J. Cleary, S. Santos, J. Henderson, D. Clark, L. Chernyak, T. A. Winningham, E. Del Barco, H. Heinrich and W. R. Buchwald, J. Opt. Soc. Am. B 25, 1708‐1713 (2008).

[Our response] Following the comment, we have added this article in the third paragraph in Section I, in the context of the propagation length of SPP.

[Reviewer 1’s Comments #10] Sec. IV, right column, spelling. “In this way, thermal annealing at 700 ◦C or **below** was found to significantly increase the grain size and reduce the surface roughness.”

[Our response] Following the comment, we have corrected the misspelling at the end of the third paragraph of Section IV.

[Reviewer 1’s Comments #11] “Additional AFM measurements confirmed that the coupler gratings had ideal rectangular profiles before and after the thermal annealing.” This result for Au (1064 C melting temperature) annealed at up to 700 C 16 min anneal differs from observations by Cleary et al. (Ref. 21) for silver (melting temperature 962 C) lamellar gratings annealed at 850 C for 30 s, which showed the rectangular profiles become more sinusoidal on annealing, significantly reducing the coupling for higher orders. It is worth commenting on the difference.

[Our response] Following the comments, we have added comments on the difference at the end of Section IV.

**Reply to Reviewer 2**

We thank Reviewer 2 for careful reading and his/her valuable comments. Our responses to the comments and the corresponding changes are described below.

**[Reviewer 2’s Comments]**

This manuscript displays interesting results regarding the effect of material properties on surface plasmon modes. The issue is clearly summarized, and the authors have been referring to the main contribution to this field throughout their paper.   
A clear correlation between the SPP losses and grain size and topology of the gold film is observed by the authors. These observations are worth publishing, as a further quantitative evidence of this correlation.   
The everlasting issue of whether the observed excess of SPP losses originate from an electrical or optical contribution is then discussed by the authors. Indeed, the as grown data might be explained by a combination of electrical (mean free path of the order of the grain size) and optical (enhanced scattering by the surface roughness, additional optical losses within electromagnetic hotspots localized at grain boundaries) contributions. The experimental evidences shown in the paper are unfortunately unable to provide their respective magnitude, but it must be reminded that this is not an easy task. The annealed datas clearly shows the interest in having a low roughness, large grain morphology to reach the promises of SPP propagation length that can be derived by ellipsometric measurements of optical index.   
The paper is logically organized, clearly written and can be published with minor corrections.

[Reviewer 2’s Comments #1] It would be more accurate to speak of "electromagnetic hotspot induced losses" rather than "SPP scattering effect" when referring to the paper of Lee. I would help to clearly differentiate their non-radiative nature (internal loss of a cavity) from the radiative nature of SPP scattering losses that can also be induced by roughness (as well described by Mills).

[Our responses] Following the comments, we have clarified non-radiative properties of the hotspot induced losses by using “electromagnetic hotspots” instead of “scattering hotspots” in the eighth paragraph of Section V. To emphasize the non-radiative properties, we also have replaced “SPP scattering” by “SPP attenuation” and “multiple SPP scattering” by “multiple SPP reflection within the hotspots” in the paragraph.

[Reviewer 2’s Comments #2] One would for example prefer the term "non radiative" instead of "irradiative" to characterize the losses.

[Our response] Following the comment, we have replaced the terms “irradiative” by “non-radiative” throughout the paper.

[Reviewer 2’s Comments #3] "evapolation" should be replace by "evaporation", at the end of the 7th paragraph of section II.

[Our response] Following the comment, we have corrected the misspelling at the end of the seventh paragraph of section II.