**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 “here we note that…the Ohmic loss” in the middle of the fourth paragraph, to present that non-radiative damping of SPP is termed as the Ohmic loss.

II. Device Design and Fabrication

* We have added “the real part of the complex” just after Eqn.(1) in the third paragraph.
* 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 the third paragraph.
* We have added four sentences at the end of Section IV, to comment on the difference between our observation and the observation by Cleary et al.

V. Discussions

* A paragraph has been added as the second paragraph, to show how the dielectric constant depends on the Drude parameters.
* The expression for the wavenumber of SPP has been corrected just after Eqn.(3) in the third paragraph: (/2) has been replaced by (/c) and tilde has been added on top of .
* A paragraph has been added as the fourth paragraph, to show the approximated form of LSPP, expressed by the dielectric constant.
* Several sentences have been added/modified in the fifth paragraph, to introduce the findings of [Trollmann et al., JPCC 2014] and to interpret our experimental results adequately.
* A paragraph has been added as the sixth paragraph, to show the derivative of LSPP with respect to the electron scattering rate , and to explain the underlying physics.
* In the seventh paragraph, we have replaced “scattering hotspots” by “electromagnetic hotspots,” “multiple SPP scattering” by “multiple SPP reflection within the hotspots,” and “SPP scattering” by “SPP attenuation.”
* Three equations (Eqn.(2), (4), (5)) have been added. Correspondingly, Eqn.(2) in the original submission has been renumbered as Eqn.(3).

Other changes

* The order of the authors have been changed (the first author and the corresponding author have not 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 of Ref. 5 and Ref.22.

**Reply to Reviewer 1**

**[Reviewer 1’s Comment #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 Comment #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 causes the relaxation of collective oscillation of free electrons, and therefore damps SPPs. Such non-radiative damping of SPPs should end up with the Joule heating, and therefore it is often termed as the Ohmic loss. Therefore the measure of the Ohmic loss is the SPP propagation length LSPP itself, determined by the non-radiative damping. In this regard, the Ohmic loss is related to the complex permittivity as expressed by Eqn.(3).

In order to display this connection more clearly, we have added Eqn.(4) in Section V. Furthermore, we have added Eqns.(2) and (5) in Section V to show how LSPP depends on the electron scattering rate .

**[Reviewer 1’s Comment #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 proportional to ’, regardless of the frequency . The cross over is important because the sign of the change in ’ upon decreasing  depends on whether  is higher than  or not. Note that ’ is proportional to ”(). In fact, *d*’()/*d* > 0 holds for  >  and *d*’()/*d* < 0 holds for  < .

Here we have added a few sentences to explain that  > holds (the operating frequency is higher than the cross over frequency) in our experiments, referring to the data of [Trollmann et al. JPCC 2014], in the fourth paragraph of Section V.

**[Our comments related to Reviewer 1’s Comment #2 and #3]**

Here we would like to explain what determines the total amount of the Ohmic loss. Although the local Joule heating efficiency is proportional to ”, the total Ohmic loss of 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 (outside metal), the total Ohmic loss becomes small, even if the local Joule-heating efficiency is high. This is why SPP’s propagate longer at terahertz and mid-infrared range than at near-infrared and visible range. Such physics is reflected to the analytic form of equation (4), which shows that LSPP depends not only on ” but also on ’2.

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 of the underlying physics are the suppression of the local Joule heating efficiency (suppression of ’) and the increase in the energy portion contained in air. The former can be checked by calculating dLSPP/d from Eqn.(4). The latter can be checked by calculating the penetration length into air and that into gold for different .

A paragraph has been added as the fourth paragraph in Section V, to give a formula of Eqn.(4), which clearly displays how LSPP depends on the dielectric constant.

Another paragraph has been added as the sixth paragraph in Section V, to show that *L*SPP increases with decreasing  and to explain the underlying physics.

**[Reviewer 1’s Comment #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.*

Theoretical consideration based on Eqn.(4, 5) tells that LSPP increases with decreasing . Our experiments showed that thermal annealing increased the grain size. What is left for connecting thermal annealing to elongation in LSPP is the correlation between the grain size and the electron scattering rate . Therefore we believe that we should show this correlation, rather than the correlation between annealing temperature and resistivity.

We have added several sentences in the fifth paragraph in Section V to introduce the experimental data which shows the correlation between the grain size and the electron scattering rate, reported in [Trollmann et al. JPCC 2014].

Some minor considerations:

**[Reviewer 1’s Comment #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 the comment, we have corrected the misspelling at the end of the 6th paragraph of Section II.

**[Reviewer 1’s Comment #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 Comment #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 Comment #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 Comment #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 cited this article at the beginning of the third paragraph in Section I.

**[Reviewer 1’s Comment #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 four sentences to comment on the difference at the end of Section IV.

**Reply to Reviewer 2**

**[Reviewer 2’s Comment #1]**

*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.*

**[Our responses]** 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 Comment #2] 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 comment, we have replaced “scattering hotspots” by “electromagnetic hotspots” in the seventh paragraph of Section V. To emphasize the non-radiative nature, we also have replaced “SPP scattering” by “SPP attenuation” and “multiple SPP scattering” by “multiple SPP reflection within the hotspots” in the same paragraph.

**[Reviewer 2’s Comment #3]** *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 Comment #4]** *"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.