Temperature dependence of THz generation from plasmonic structures

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Abstract

We investigate the efficiency of optically-induced THz generation from plasmonic structures as a function of temperature. THz generation from plasmonic structures is critically dependent on electromagnetic field enhancement, with high order dependence on incident optical intensity. Thus, THz generation from plasmonic structures is extremely sensitive to changes in the local field enhancement. Over a range of temperatures from 300 K down to 10 K we observe 6-fold enhancement of the generated THz intensity. We suggest this effect is most likely due to a reduction the ohmic losses resulting from diminished electron-phonon scattering.

1. Introduction

The field of plasmonics relies on the collective oscillations of free electrons in metals. All the plasmonic related effects suffer from the strong damping of the electron oscillation which is intrinsic property of normal metals. The effects of plasmonic sensing, plasmon-assisted electron emission, SERS and others would all benefit from a reduction of the intrinsic losses in the metal. In recent years great efforts were made to overcome the losses in plasmonics. One of the ways to alleviate SPP damping is through the use of gain media.

Whilst investigations in coupling of plasmonics and gain media shows great potential, another possibility for suppressing electron oscillation damping, namely low temperatures, has arguably received less attention. Only recently the effects of low temperatures in plasmonics were investigated [1-4] and showed the advantages of such an approach. In this paper we describe our investigation of the effect of the temperature on plasmonic oscillations by using the phenomenon of THz generation from plasmonic structures [5].

2. THz generation from plasmonic structures

We used semicontinuous silver films to generate THz radiation. The films were fabricated by evaporation of silver in vacuum onto a glass substrate at a pressure < 10^{-5} mbar and at an evaporation rate of 1 Å/s. It was shown recently that these films strongly enhance electromagnetic field [6] and may be used to generate THz radiation [7], [8]. An important feature is an observed ~ 5-6th order dependence of the THz intensity on the incident optical intensity, which indicates that the generation mechanism is not 2nd order optical rectification (as described in ref. [8]). Our observations, reported in ref. [7], are more consistent with a mechanism proposed originally by Welsh et al. [9], in which excitation of localised surface plasmons and multiphoton absorption leads to the enhancement of the electromagnetic field around the plasmonic structure and subsequent electron emission. The ejected electrons are subject to

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forces exerted on them by the electromagnetic field. In the plasmon resonance condition this is mainly the plasmon-enhanced field. The localised nature of the plasmonic field leads to a high inhomogeneity of the field, leading to a ponderomotive acceleration of the electrons. The time scale of the acceleration is determined by the spatial variation in the incident field and, together with duration of pulse (~ 100 fs), leads the generation of radiation in the THz range. The high order dependence of THz generation according to the model is a result of the multiphoton excitation at low optical intensities. Since the electrons are ejected and accelerated by the enhanced plasmonic field, the high power dependence of the THz radiation efficiency is highly sensitive to the changes in enhancement of the electromagnetic field.

3. Temperature dependence

The plasmon damping of noble metal structures originates from the electron-electron and electron-phonon scattering and radiative losses. The radiative, τ_r, and electron-electron relaxation time, $\tau_{e\text{-}e}$, were investigated in [3] and [10-12] and their temperature dependences are negligible. The electron-phonon scattering time, $\tau_{e\text{-}p}$, was discussed in [13] and [14] and is given by,

$$\tau_{e-p}^{-1}(T) = \tau_0^{-1}\left[\frac{2}{5} + 4\left(\frac{T}{\Theta}\right)^5 \int_0^{\Theta/T} \frac{z^4}{e^z - 1} dz\right], \qquad (1)$$

where τ_0 is material-dependent constant and Θ is the Debye's temperature. The damping rate is the sum of electron-electron scattering, electron-phonon scattering and radiative relaxation rate and is given by

$$\gamma = \hbar\left[\tau_{e-e}^{-1} + \tau_{e-p}^{-1} + \tau_r^{-1}\right], \qquad (2)$$

and is a function nearly linearly decreasing with temperature in the range 300 – 100 K with a saturation below 50 K [3].

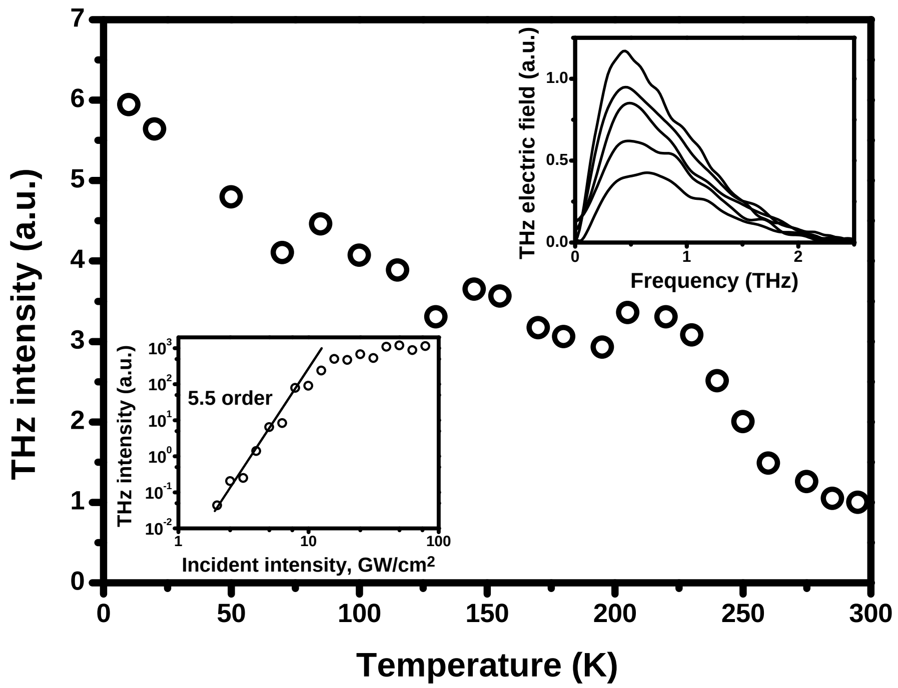

Fig. 1: Temperature dependence of the emmitted THz intensity. The intensity is normalised to the lowest value (room temp). Lower inset: Intensity dependence of THz radiation showing high-order dependence. Upper inset: THz radiation spectra at the following temperatures: 295, 250, 220, 85 and 10 K.

Reduction in the damping parameter of the plasmon resonance results in an increase of the quality factor for a plasmonic resonance, which is inversely proportional to the imaginary part of the dielectric constant [4]. To investigate the effect of temperature on THz generation efficiency we used a conventional THz spectrometer [7], the sample being placed in a cryostat. The effect was investigated in the temperature range 300 – 10 K, and the resultant dependence of the emmitted radiation intensity as a function of temperature is shown in the Fig.1. The electric field is normalised to the lowest value at room temperature and thus the maximum intensity enhancement as can be seen from the fig.1 is 6 times.

4. Conclusion

In conclusion, we observed temperature dependence of THz radiation from semicontinuous silver films in the temperature range 300 – 10 K. We assign this effect to the reduction of the SPP damping, namely the suppression of the electron-phonon scattering. The high order dependence of the emitted THz intensity on incident optical intensity could be utilized for sensitive investigations of low-temperature plasmonic effects and SPP damping mechanisms as well as the interaction of plasmonic structures with gain media.

References

1. Pang, Y.S., H.J. Hwang, and M.S. Kim, Reversible temperature dependence in surface-enhanced Raman scattering of 1-propanethiol adsorbed on a silver island film. *Journal of Physical Chemistry B*, 1998. 102(37): p. 7203-7209.
2. Ozdemir, S.K. and G. Turhan-Sayan, Temperature effects on surface plasmon resonance*:* Design considerations for an optical temperature sensor. *Journal of Lightwave Technology*, 2003. 21(3): p. 805-814.
3. Liu, M.Z., M. Pelton, and P. Guyot-Sionnest, Reduced damping of surface plasmons at low temperatures. *Physical Review B*, 2009. 79(3): p. 5.
4. Bouillard, J.S., et al., Low-temperature plasmonics of metallic nanostructures. *Nano Lett*, 2012. 12(3): p. 1561-5.
5. Kadlec, F., P. Kuzel, and J.L. Coutaz, Optical rectification at metal surfaces. *Optics Letters,* 2004. 29(22): p. 2674-2676.
6. Chettiar, U.K., et al., FDTD modeling of realistic semicontinuous metal films. *Applied Physics B-Lasers and Optics*, 2010. 100(1): p. 159-168.
7. Polyushkin, D.K., et al., THz generation from plasmonic nanoparticle arrays. *Nano Lett,* 2011. 11(11): p. 4718-24.
8. Ramakrishnan, G. and P.C.M. Planken, Percolation-enhanced generation of terahertz pulses by optical rectification on ultrathin gold films. *Optics Letters,* 2011. 36(13): p. 2572-2574.
9. Welsh, G.H., N.T. Hunt, and K. Wynne, Terahertz-pulse emission through laser excitation of surface plasmons in a metal grating. *Physical Review Letters,* 2007. 98(2): p. 4.
10. Gurzhi, R.N., MUTUAL ELECTRON CORRELATIONS IN METAL OPTICS. *Soviet Physics Jetp-Ussr,* 1959. 8(4): p. 673-675.
11. Lawrence, W.E. and J.W. Wilkins, Electron-Electron Scattering in the Transport Coefficients of Simple Metals. *Physical Review B,* 1973. 7(6): p. 2317-2332.
12. Parkins, G.R., W.E. Lawrence, and R.W. Christy, Intraband optical conductivity σ(ω,T) of Cu, Ag, and Au: Contribution from electron-electron scattering. *Physical Review B,* 1981. 23(12): p. 6408-6416.
13. Gurzhi, R.N., Sov. Phys. JETP, 1958. 6.
14. Holstein, T., Theory of transport phenomena in an electron-phonon gas. *Ann. Phys.,* 1964. 29.

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