

Polarized thermal emission from metamaterials described by Stokes parameters based on fluctuational electrodynamics

Zhuomin Zhang and Chiyu Yang

G. W. Woodruff School of Mechanical Engineering Georgia Institute of Technology, Atlanta, Georgia

Quantum and Thermal Electrodynamic Fluctuations in the Presence of Matter: Progress and Challenges July 11–14, 2022 KITP, UC Santa Barbara



Emerging Regimes and Implications of Quantum and Thermal Fluctuational Electrodynamics June 20-August 4, 2022



Outline

- Introduction and overview
- Interpretation of Kirchhoff's law
- Fluctuational electrodynamics and Stokes' parameters
- Computational results
- Conclusions & Acknowledgments

Micro/Nanoscale Thermal Radiation

- Micro/nanoscale thermal radiation concerns both near-field radiative heat transfer (NFRHT) between closely spaced objects and the interaction of electromagnetic waves with micro/nanostructured materials that could potentially modify the far-field radiative properties.
- Examples of micro/nanostructures, gratings, nanowires, nanotubes, multilayers, nanoparticles and clusters, graphene and 2D van der Waals materials, graphene ribbons, magneto-optical materials, etc.

Optical Metamaterials with Micro/Nanostructures



1D photonic crystals/hyperbolic meta Appl. Phys. Lett. **87**, 071904 (2005) J. Appl. Phys. **100**, 063529 (2006) Opt. Lett. **33**, 204 (2008) JQSRT **197**, 132 (2017)



Grating multilayered metamaterials Opt. Express **16**, 11328 (2008) Phys. Rev. E **84**, 026603 (2011) Appl. Phys. Lett. **100**, 063902 (2012) Int. J. Heat Mass Transfer (2013)



Si nanowire metamaterials

J. Heat Transfer **135**, 061602 (2013) *Appl. Phys. Lett.* **103**, 103101 (2013) *Int. I. Thermo. Sci.* **65**, 62 (2013)



Carbon nanotube arrays Nanotechnology **20**, 215704 (2009) Appl. Phys. Lett. **97**, 163116 (2010) Appl. Phys. Lett. **101**, 141909 (2012)



Complex gratings

Opt. Commun. **269**, 411 (2007) *JOSA_B* **27**, 2595 (2010) *Opt. Express* **21**, 10502 (2013)



Ag nanorod arrays Appl. Opt. **51**, 1521 (2012) NMTE **16**, 18 (2012) ARHT **16**, 351 (2013)

2D Materials Coupled with Nanostructures



Appl. Phys. Lett. **105**, 031905 (2014) JOSA_B **32**, 1176 (2015) ACS Photon. **2**, 1611 (2015) J. Opt. **17**, 035004 (2015) Appl. Phys. Lett. **107**, 191112 (2015)



J. Heat Transfer **139**, 022701 (2017) *IJHMT* **106**, 1025 (2017) *MNTE* **21**, 123 (2017) *Opt. Express* **25**, 7791 (2017) *Phys. Rev. B* **95**, 245437 (2017) *Solar Energy* **159**, 329 (2018)

Hexagonal boron-nitride (hBN)

Bi₂Te₃

Record-High Solar Reflectance for Daytime Radiative Cooling



Yang et al. (2018), Solar Energy 169, 316

Near-Field Radiative Heat Transfer



Also hBN, MoO₃, etc.

Our Measurement Results



Watjen, Zhao, and Zhang, Appl. Phys. Lett. 109, 203112 (2016).

Outline

- Introduction and overview
- Interpretation of Kirchhoff's law
- Fluctuational electrodynamics and Stokes' parameters
- Computational results
- Conclusions & Acknowledgments

Conventional Kirchhoff's Law

Choose a particular direction (θ , ϕ), let the absorbed power equal to the incident power:



Absorbed: $I_{b\lambda}(\lambda, T_w)\alpha(\lambda, \theta, \phi)\cos\theta d\Omega_i dA_1$ Emitted: $I_{b\lambda}(\lambda, T_s)\varepsilon(\lambda, \theta, \phi)\cos\theta d\Omega_e dA_1$

For the same solid angle and area when $T_s = T_w$ (thermal equilibrium), we have:

$$\alpha_{\lambda}(\lambda,\theta,\phi) = \varepsilon_{\lambda}(\lambda,\theta,\phi)$$

"Conventional" Kirchhoff's law.

This equality is NOT necessary since it is not required by the energy balance. Nevertheless, this equation always works for reciprocal materials (most of the materials we deal with).

A More Proper Formulism



Note that the outgoing radiation is made up by the emitted and reflected.

We know that the incident radiation intensity is blackbody distribution regardless of the direction: $I_{b\lambda}(\lambda,T)$

The outgoing radiation towards (θ_r, ϕ_r) consists of emitted and reflected. However, the combined intensity must also be uniform and equal to the blackbody intensity: $I_{outgoing} = I_{b\lambda}(\lambda, T)$

Assume specular surface (smooth and mirror reflection without transmission):

 $\varepsilon(\lambda, \theta_{\mathrm{r}}, \phi_{\mathrm{r}}) + \rho(\lambda, \theta_{\mathrm{i}}, \phi_{\mathrm{i}}) = 1 \text{ and } \alpha(\lambda, \theta_{\mathrm{i}}, \phi_{\mathrm{i}}) = 1 - \rho(\lambda, \theta_{\mathrm{i}}, \phi_{\mathrm{i}})$

 $\implies \varepsilon(\lambda, \theta_{\rm r}, \phi_{\rm r}) = \alpha(\lambda, \theta_{\rm i}, \phi_{\rm i})$

Reciprocal Materials



 $\rho(\lambda, \theta_{i}, \phi_{i}) = \rho(\lambda, \theta_{r}, \phi_{r})$ (symmetry)

 $\alpha(\lambda, \theta_{i}, \phi_{i}) = \alpha(\lambda, \theta_{r}, \phi_{r}) \quad \text{(symmetry)}$

 $\varepsilon(\lambda, \theta_{i}, \phi_{i}) = \alpha(\lambda, \theta_{i}, \phi_{i}) = \alpha(\lambda, \theta_{r}, \phi_{r})$

Key points:

(1) $\varepsilon(\lambda, \theta_i, \phi_i) = \alpha(\lambda, \theta_r, \phi_r)$ General material (flat, opaque) (2) $\varepsilon(\lambda, \theta_i, \phi_i) = \alpha(\lambda, \theta_i, \phi_i)$ Reciprocal materials only

So far, we talked about absorptivity is for randomly polarized irradiation; hence, the radiative properties are for averaged polarizations:

$$\varepsilon = \frac{\varepsilon_p + \varepsilon_s}{2}$$
 and $\alpha = \frac{\alpha_p + \alpha_s}{2}$

Polarization Dependence

Co-polarization and cross-polarization: $\rho_p = \rho_{pp} + \rho_{ps}$ and $\rho_s = \rho_{sp} + \rho_{ss}$

Use *p*-pol as an example (specular reflection only).

$$\alpha_{p}(\theta_{i},\phi_{i}) = 1 - \rho_{p}(\theta_{i},\phi_{i})$$

$$\alpha_{p}^{(i)} = 1 - \rho_{pp}^{(i)} - \rho_{ps}^{(i)}$$

$$\varepsilon_{p}^{(r)} + \rho_{pp}^{(i)} + \rho_{sp}^{(i)} = 1$$

$$\varepsilon_{p}^{(r)} = 1 - \rho_{pp}^{(i)} - \rho_{sp}^{(i)}$$

If no cross-polarization ($\rho_{sp} = 0 \& \rho_{ps} = 0$): $\alpha_p^{(i)} = \varepsilon_p^{(r)}$

For both reciprocal and nonreciprocal

If reciprocal (
$$\rho_{pp}^{(i)} = \rho_{pp}^{(r)} \& \rho_{sp}^{(i)} = \rho_{ps}^{(r)}$$
): $\alpha_p^{(r)} = \varepsilon_p^{(r)}$ and $\alpha_p^{(i)} = \varepsilon_p^{(i)}$

Zhang et al. (2020), JQSRT **245**, 106904.

Regime Map



No diffraction effects. No rough surface scattering. Opaque.

Outline

- Introduction and overview
- Interpretation of Kirchhoff's law
- Fluctuational electrodynamics and Stokes' parameters
- Computational results
- Conclusions & Acknowledgments

Fluctuation-Dissipation Theorem

The polarization status of thermal radiation



The materials considered may be anisotropic and nonreciprocal, and may contain multiple layers but each layer is homogeneous. Thermal emission is largely unpolarized or randomly polarized !

Can thermal emission be circularly polarized?

How to model thermal emission?

Fundamentally, thermal emission is due to charge fluctuations. Here, we use fluctuation-dissipation theorem coupled with dyadic Green's functions to model the emitted electromagnetic (EM) field.

From the EM field, we determine Stokes' parameters to fully characterize the intensity and polarization status.

Stokes' Parameters

$$\mathbf{S} = \begin{pmatrix} S_0 \\ S_1 \\ S_2 \\ S_3 \end{pmatrix} = \begin{pmatrix} I_{0^0} + I_{90^\circ} \\ I_{0^0} - I_{90^\circ} \\ I_{45^0} - I_{-45^\circ} \\ I_{R} - I_{L} \end{pmatrix}$$

I, S: Optical intensity for different polarizations

$$\begin{split} S_0 &= I_{\text{tot}} = I_{0^0} + I_{90^\circ} = I_{45^0} + I_{-45^\circ} = I_{\text{R}} + I_{\text{L}} \\ S_0^2 &\geq S_1^2 + S_2^2 + S_3^2 = I_{\text{pol}}^2 \end{split}$$

Degree of polarization (DoP):

$$P = \frac{\sqrt{S_1^2 + S_2^2 + S_3^2}}{S_0} = \frac{I_{\text{pol}}}{I_{\text{tot}}}; \quad P_1 = \frac{\sqrt{S_1^2 + S_2^2}}{S_0}; \quad P_c = \frac{|S_3|}{S_0}$$

One may split a partially polarized beam to two parts: one is unpolarized and the other is completely polarized such that

$$\mathbf{S} = \begin{pmatrix} S_0 \\ S_1 \\ S_2 \\ S_3 \end{pmatrix} = \begin{pmatrix} S_{unp} \\ 0 \\ 0 \\ 0 \end{pmatrix} + \begin{pmatrix} S_{pol} \\ S_1 \\ S_2 \\ S_3 \end{pmatrix} = \begin{pmatrix} (1-P)S_0 \\ 0 \\ 0 \\ 0 \end{pmatrix} + \begin{pmatrix} S_{pol} \\ S_1 \\ S_2 \\ 0 \end{pmatrix}$$

Stokes' vector **S** and parameters, $S_{0,1,2,3}$.

Electric Field and Emissivity Vector

$$\begin{bmatrix} S_{0} \\ S_{1} \\ S_{2} \\ S_{3} \end{bmatrix} = \begin{bmatrix} \langle E_{x}E_{x}^{*} \rangle + \langle E_{y}E_{y}^{*} \rangle \\ \langle E_{x}E_{x}^{*} \rangle - \langle E_{y}E_{y}^{*} \rangle \\ \langle E_{x}E_{y}^{*} \rangle + \langle E_{y}E_{x}^{*} \rangle \\ i \langle E_{x}E_{y}^{*} \rangle - i \langle E_{y}E_{x}^{*} \rangle \end{bmatrix} \xrightarrow{Equivalent} \begin{bmatrix} \langle E_{x}E_{y}^{*} \rangle \\ \langle E_{x}E_{y}^{*} \rangle \\ i \langle E_{x}E_{y}^{*} \rangle + \langle E_{y}E_{x}^{*} \rangle \end{bmatrix}$$

$$= \begin{bmatrix} \left\langle E_p E_p^* \right\rangle + \left\langle E_s E_s^* \right\rangle \\ \left\langle E_p E_p^* \right\rangle - \left\langle E_s E_s^* \right\rangle \\ \left\langle E_p E_s^* \right\rangle + \left\langle E_p E_s^* \right\rangle \\ i \left\langle E_p E_s^* \right\rangle - i \left\langle E_p E_s^* \right\rangle \end{bmatrix} = \begin{bmatrix} I_{0^0} + I_{90^\circ} \\ I_{0^0} - I_{90^\circ} \\ I_{45^0} - I_{-45^\circ} \\ I_R - I_L \end{bmatrix}$$

Here, E_p and E_s are the field components normal to the propagation direction.

Thermal emission and emissivities:

$$\begin{bmatrix} S_0 \\ S_1 \\ S_2 \\ S_3 \end{bmatrix} = I_{bb,\theta} \begin{bmatrix} \epsilon_{0^\circ} + \epsilon_{90^\circ} \\ \epsilon_{0^\circ} - \epsilon_{90^\circ} \\ \epsilon_{45^\circ} - \epsilon_{-45^\circ} \\ \epsilon_{R} - \epsilon_{L} \end{bmatrix}$$

Here, $I_{bb,\theta}$ is the (spectral) optical intensity of a blackbody that depends on the emission polar angle θ .

The next question is: How to calculate the electrical field of thermal emission?

Fluctuational Electrodynamics Formulism

$$\mathbf{E}(\mathbf{r},\omega) = \mathrm{i}\omega\mu_0 \int \vec{G}(\mathbf{r},\mathbf{r}',\omega) \cdot \mathbf{J}(\mathbf{r}',\omega) \mathrm{d}\mathbf{r}'$$
$$\mathbf{E}(\mathbf{r}_1,\omega) \otimes \mathbf{E}^*(\mathbf{r}_2,\omega) \Big\rangle = \Big\langle \mu_0^2 \omega^2 \int \int \vec{G}(\mathbf{r}_1,\mathbf{r}',\omega) \mathbf{J}(\mathbf{r}',\omega) \mathbf{J}^{\dagger}(\mathbf{r}'',\omega) \vec{G}^{\dagger}(\mathbf{r}_2,\mathbf{r}'',\omega) \mathrm{d}\mathbf{r}' \mathrm{d}\mathbf{r}'' \Big\rangle$$
$$\Big\langle \mathbf{E}(\mathbf{r},\omega) \otimes \mathbf{E}^*(\mathbf{r},\omega) \Big\rangle = \begin{bmatrix} \Big\langle E_x E_x^* \Big\rangle & \big\langle E_x E_y^* \big\rangle & \big\langle E_x E_z^* \big\rangle \\ \big\langle E_y E_x^* \big\rangle & \big\langle E_y E_y^* \big\rangle & \big\langle E_y E_z^* \big\rangle \\ \big\langle E_z E_x^* \big\rangle & \big\langle E_z E_y^* \big\rangle & \big\langle E_z E_z^* \big\rangle \end{bmatrix} \mathbf{3} \times \mathbf{3} \text{ matrix}$$

For a nonmagnetic material.

$$\left\langle \mathbf{J}(\mathbf{r}',\omega) \otimes \mathbf{J}^{*}(\mathbf{r}'',\omega) \right\rangle = \frac{4}{\pi} \omega \varepsilon_{0} \Theta(\omega,T) \frac{\vec{\varepsilon}(\omega) - \vec{\varepsilon}^{\dagger}(\omega)}{2\mathrm{i}} \delta(\mathbf{r}' - \mathbf{r}'')$$

Conversion to wavevector integration:

$$\mathbf{E}(\mathbf{r},\omega) = \int \frac{d\mathbf{k}_{\rho}}{4\pi^2} \mathbf{E}(z,\omega,\mathbf{k}_{\rho}) e^{i\mathbf{R}\cdot\mathbf{k}_{\rho}}$$
$$\mathbf{R} = x\hat{\mathbf{x}} + y\hat{\mathbf{y}}; \ \mathbf{k}_{\rho} = k_x\hat{\mathbf{x}} + k_y\hat{\mathbf{y}}; \ \text{and} \ k_z = \sqrt{k_0^2 - \mathbf{k}_{\rho}\cdot\mathbf{k}_{\rho}} = \sqrt{k_0^2 - k_{\rho}^2}$$

Wavevector Space

Scattering theory:
$$\frac{\omega^2}{c^2} \int \vec{G}(\mathbf{r}_1, \mathbf{r}', \omega) \frac{\left[\vec{\varepsilon}(\omega) - \vec{\varepsilon}^{\dagger}(\omega)\right]}{2i} \vec{G}^{\dagger}(\mathbf{r}_2, \mathbf{r}', \omega) d\mathbf{r}'$$
$$= \frac{\vec{G}(\mathbf{r}_1, \mathbf{r}_2, \omega) - \vec{G}^{\dagger}(\mathbf{r}_2, \mathbf{r}_1, \omega)}{2i}$$

Conversion to \mathbf{k}_{ρ} Space:

$$\vec{G}(\mathbf{r}_1, \mathbf{r}_2, \omega) = \int \frac{\mathrm{d}\mathbf{k}_{\rho}}{4\pi^2} \vec{g}(z_1, z_2, \mathbf{k}_{\rho}, \omega) \mathrm{e}^{\mathrm{i}\mathbf{k}_{\rho} \cdot (\mathbf{R}_1 - \mathbf{R}_2)}$$

Here, \vec{g} is obtained based on reflection coefficients.

Results:

$$\left\langle \mathbf{E}(z,\omega,\mathbf{k}_{\rho}) \otimes \mathbf{E}^{*}(z,\omega,\mathbf{k}_{\rho}) \right\rangle = \begin{pmatrix} \left\langle E_{x}E_{x}^{*} \right\rangle & \left\langle E_{x}E_{y}^{*} \right\rangle & \left\langle E_{x}E_{z}^{*} \right\rangle \\ \left\langle E_{y}E_{x}^{*} \right\rangle & \left\langle E_{y}E_{y}^{*} \right\rangle & \left\langle E_{y}E_{z}^{*} \right\rangle \\ \left\langle E_{z}E_{x}^{*} \right\rangle & \left\langle E_{z}E_{y}^{*} \right\rangle & \left\langle E_{z}E_{z}^{*} \right\rangle \end{pmatrix}_{z,\omega,\mathbf{k}_{\rho}}$$

If $k_{\rho} \le k_0$, propagating waves only, far-field results (independent of *z*). By coordinates transformation, we can set the *z*'-direction to the **k**-direction and then evaluate the 2 x 2 matrix with $E_{x'}$ and $E_{y'}$ only.

Outline

- Introduction and overview
- Interpretation of Kirchhoff's law
- Fluctuational electrodynamics and Stokes' parameters
- Computational results
- Conclusions & Acknowledgments

Structure #1: Hexagonal boron nitride (hBN)



Two layers of hBN films with in-plane optical axis; Top layer rotated in order to get circular polarization.

Double-Layer hBN on Glass



 $\lambda = 6.3 \ \mu m$ $\theta = 52^{\circ}$

Tunable linear or circular polarization with DoP greater than 0.75 !

The structure is reciprocal according to conventional Kirchhoff's law but not symmetry with respect to $\phi = 90^{\circ}$.

Polarization Ellipse



Wavelength Dependence



hBN films on glass Normal direction:

$$heta=0^\circ$$
, $\phi=0^\circ$

When the difference between orthogonal polarizations is small, it tends to be randomly polarized. The (average) emissivity could approach 1.

When the DoP is 1 or 100% polarized, the (polarization-averaged) emissivity is 0.5 since the orthogonal component is zero.

Weyl Semimetal (WSM) – Nonreciprocal Materials



$$\vec{\varepsilon} = \begin{bmatrix} \varepsilon_{d} & i\varepsilon_{a} & 0\\ -i\varepsilon_{a} & \varepsilon_{d} & 0\\ 0 & 0 & \varepsilon_{d} \end{bmatrix} \qquad \begin{array}{c} \vec{\varepsilon}^{\mathrm{T}} \neq \vec{\varepsilon} \\ \text{(nonreciprocal)} \end{array}$$

Parameters from

Yang et al. (2022) Opt. Express 30, 3035



Note that **b** is a vector and if the direction of b is changed, the dielectric tensor will be transposed. Similar to magneto-optical materials with an external magnetic field.

Structure #2 Bulk Weyl Semimetal (WSM)



Nanoscale Thermal Radiation Lab

27

Structure #3 WSM + hBN Multilayer







Absorptivity p,s,R,L polarization

Emissivity p,s,R,L polarization

Kirchhoff's law is not valid for any polarization !

(General) Kirchhoff's Law





Structure #3 at λ = 7.6 µm

Adjoint Kirchhoff's Law



For the same conditions as before.

(Reversed Weyl **b** for this case)

See paper from Prof. Shanhui Fan's group, Guo et al. PRX (2022).

Structure #3 Azimuthal Angle Dependence



No apparent symmetry; contains random, linear, and circular polarizations

Structure #3 Polar Angle Dependence



Nanoscale Thermal Radiation Lab

32

Conclusions

- Fluctuational electrodynamics is used to derive Stokes' parameters for thermal emission from layered anisotropic media, including nonreciprocal materials to fully characterize the polarization status of thermal emission.
- Thermal emission from anisotropic materials may be circularly polarized as well as linearly polarized. This study will help design ideal thermal emitter for energy harvesting and thermal control.
- The general Kirchhoff's law and adjoint Kirchhoff's law are validated for nonreciprocal materials. For reciprocal materials, the conventional Kirchhoff's law always holds for each individual polarization.

Acknowledgements

National Science Foundation (NSF)

Grant No. CBET-2029892; Grant No. PHY-1748958 (KITP Program).

All collaborators over the years, and fruitful discussion with colleagues at the KITP FLECTRO22 program

http://zhang-nano.me.gatech.edu/