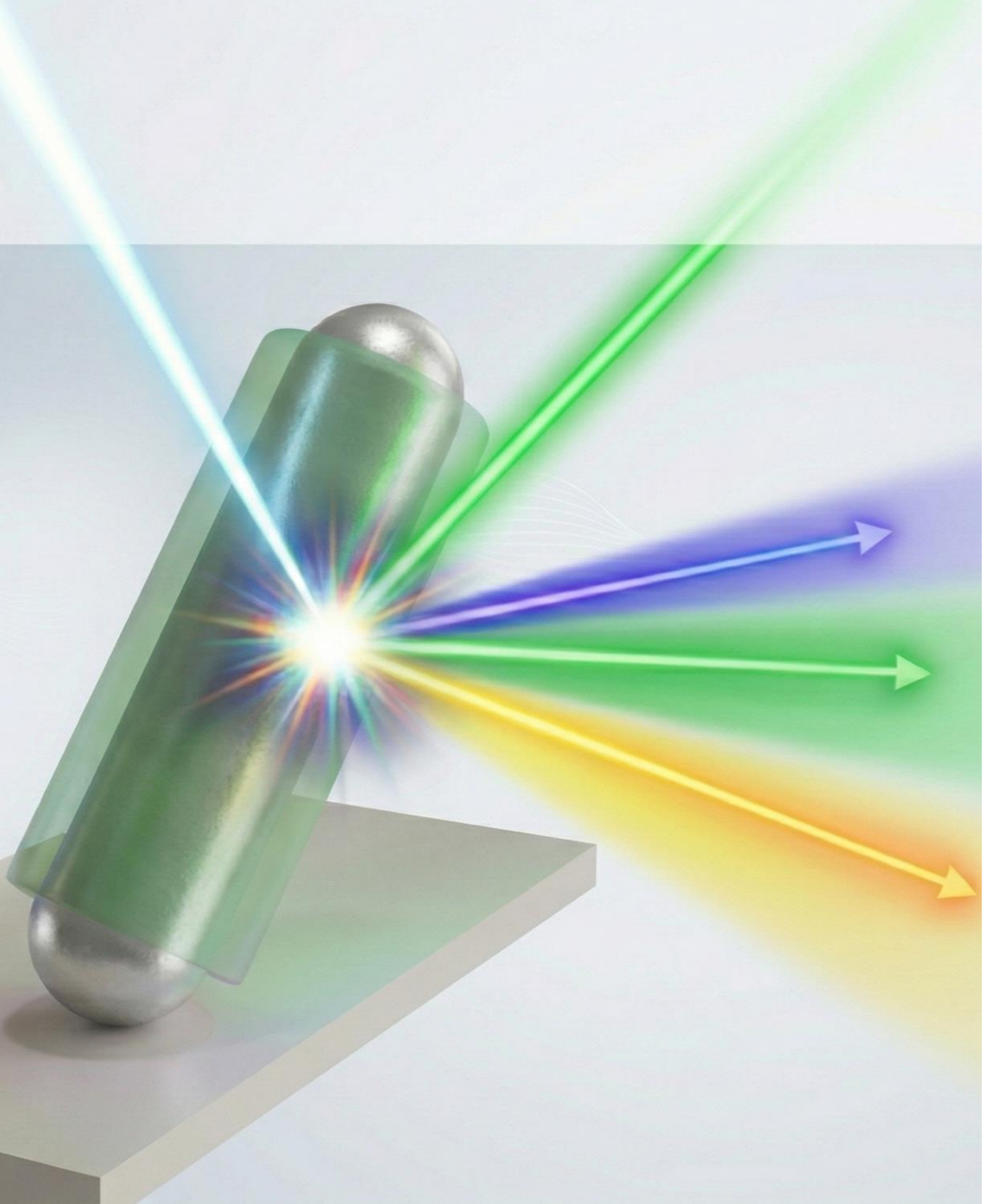

Probing low-frequency excitations with visible light and electron beams

Leila Prelat

ICFO-The Institute of Photonic Sciences, Barcelona, Spain

Nanolight Benasque 2026

nanophotonics.org

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Technological applications of nonlinear optics

Laser technology

Parametric amplifiers

Quantum optics

Nonlinear optics

Sensing

Signal processing

Telecom

Nonlinear optics **provides insights** into the behavior of optical fields, going beyond the linear response of materials

Most materials present **weak nonlinear response**, thus finding ways to enhance it is an important challenge

Linear optics

$$\mathbf{P}(t) \propto \chi^{(1)} \mathbf{E}(t)$$

Nonlinear optics

$$\mathbf{P}(t) \propto \chi^{(1)}\mathbf{E}(t) + \chi^{(2)}\mathbf{E}^2(t) + \chi^{(3)}\mathbf{E}^3(t) + \ldots$$

Nonlinear optics: second order

$$E(t) = E_1\,\mathrm{e}^{-\mathrm{i}\omega_1 t} + E_2\,\mathrm{e}^{-\mathrm{i}\omega_2 t} + \text{c.c.}$$

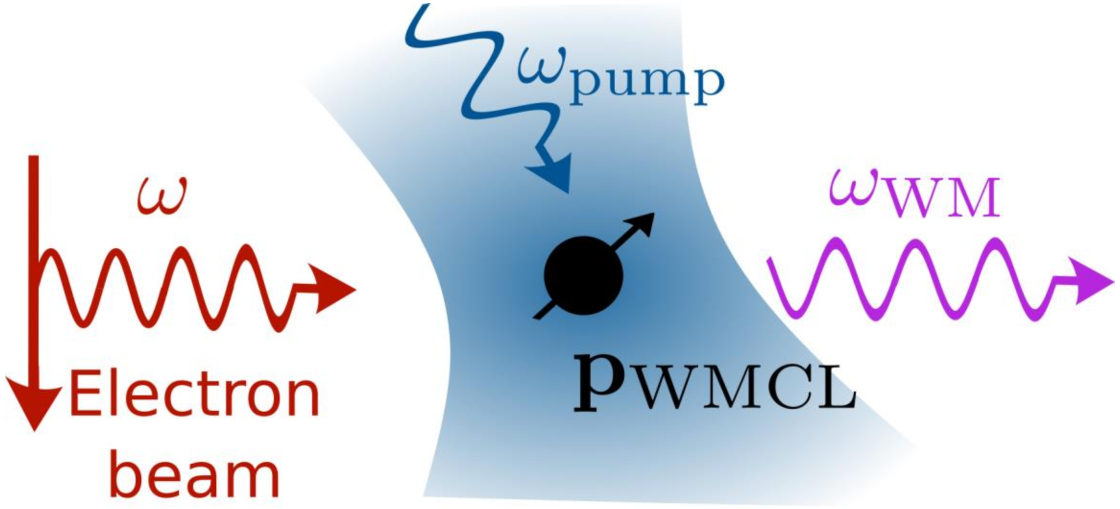
$$P^{(2)}(t) \propto \chi^{(2)}\Big[E_1^2\,\mathrm{e}^{-2\mathrm{i}\omega_1 t} + E_2^2\,\mathrm{e}^{-2\mathrm{i}\omega_2 t} + \overset{\text{SFG}}{\boxed{2E_1E_2\,\mathrm{e}^{-\mathrm{i}(\omega_1+\omega_2)t}}} +$$

$$+ \overset{\text{DFG}}{\boxed{2E_1E_2^*\mathrm{e}^{-\mathrm{i}(\omega_1-\omega_2)t}}} + E_1E_1^* + E_2E_2^* + \text{c.c.}\Big]$$

Emission rate of output photons for an electrostatic cavity

Induced dipole moment:

$$\mathbf{p}_{\mathrm{WMCL}}(\omega_{\mathrm{WM}}) = \chi^{(2)}(\omega_{\mathrm{pump}}, \omega, \omega_{\mathrm{WM}}) : \mathbf{E}^{\mathrm{ind}}_{\mathrm{pump}}(\omega_{\mathrm{pump}})\mathbf{E}^{\mathrm{ind}}_{\mathrm{electron}}(\omega)$$



Emission rate of output photons for an electrostatic cavity

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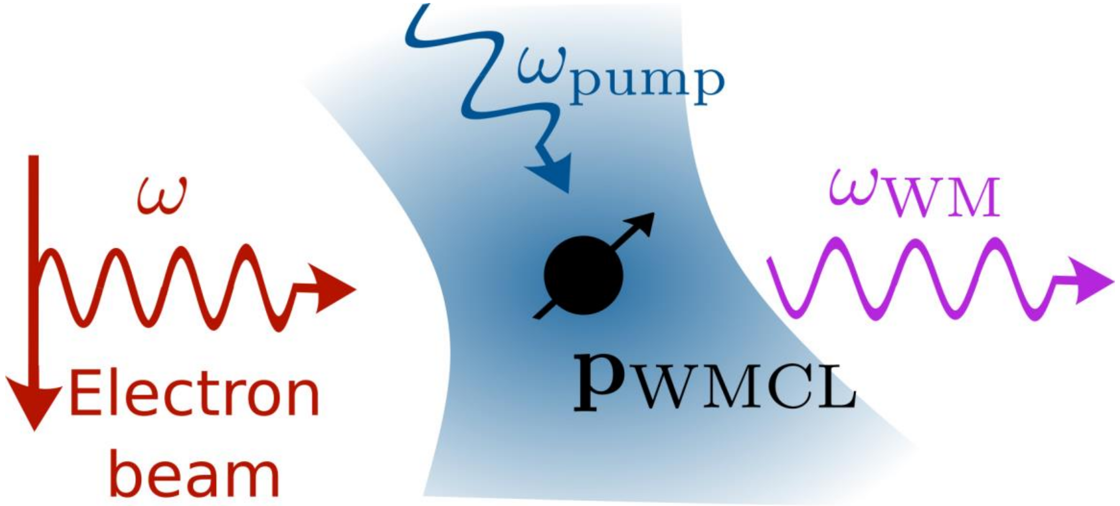
$$\mathbf{p}_{\mathrm{WMCL}}(\omega_{\mathrm{WM}}) = \chi^{(2)}(\omega_{\mathrm{pump}}, \omega, \omega_{\mathrm{WM}}) : \boxed{\mathbf{E}^{\mathrm{ind}}_{\mathrm{pump}}(\omega_{\mathrm{pump}})\mathbf{E}^{\mathrm{ind}}_{\mathrm{electron}}(\omega)}$$

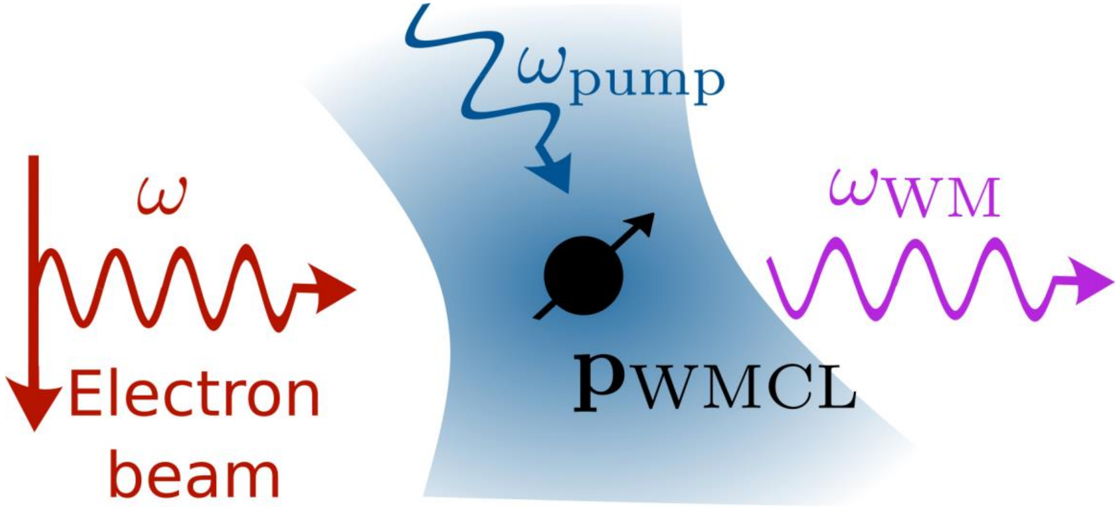


$$\mathbf{E}^{\mathrm{ind}}(\omega) = \frac{3}{\epsilon(\omega)+2}\,\mathbf{E}^{\mathrm{ext}}(\omega)$$

Emission rate of output photons for an electrostatic cavity

Induced dipole moment:

$$\mathbf{p}_{\mathrm{WMCL}}(\omega_{\mathrm{WM}}) = \chi^{(2)}(\omega_{\mathrm{pump}}, \omega, \omega_{\mathrm{WM}}) : \mathbf{E}^{\mathrm{ind}}_{\mathrm{pump}}(\omega_{\mathrm{pump}})\mathbf{E}^{\mathrm{ind}}_{\mathrm{electron}}(\omega)$$



$$\mathbf{E}^{\mathrm{ind}}(\omega) = \frac{3}{\epsilon(\omega) + 2}\mathbf{E}^{\mathrm{ext}}(\omega)$$

Emission rate of output photons:

$$\Gamma_{\mathrm{WM}}(\omega_{\mathrm{WM}}) \propto \frac{\omega_{\mathrm{WM}}^3 \omega^2}{c^3 v^4 \gamma^2} K_1^2\left(\frac{\omega b}{v\gamma}\right) \frac{|\mathbf{E}_{\mathrm{pump}}|^2 |\chi^{(2)}|^2}{|[\epsilon(\omega_{\mathrm{pump}}) + 2][\epsilon(\omega_{\mathrm{WM}}) + 2][\epsilon(\omega) + 2]|^2}$$

Emission rate of output photons for an electrostatic cavity

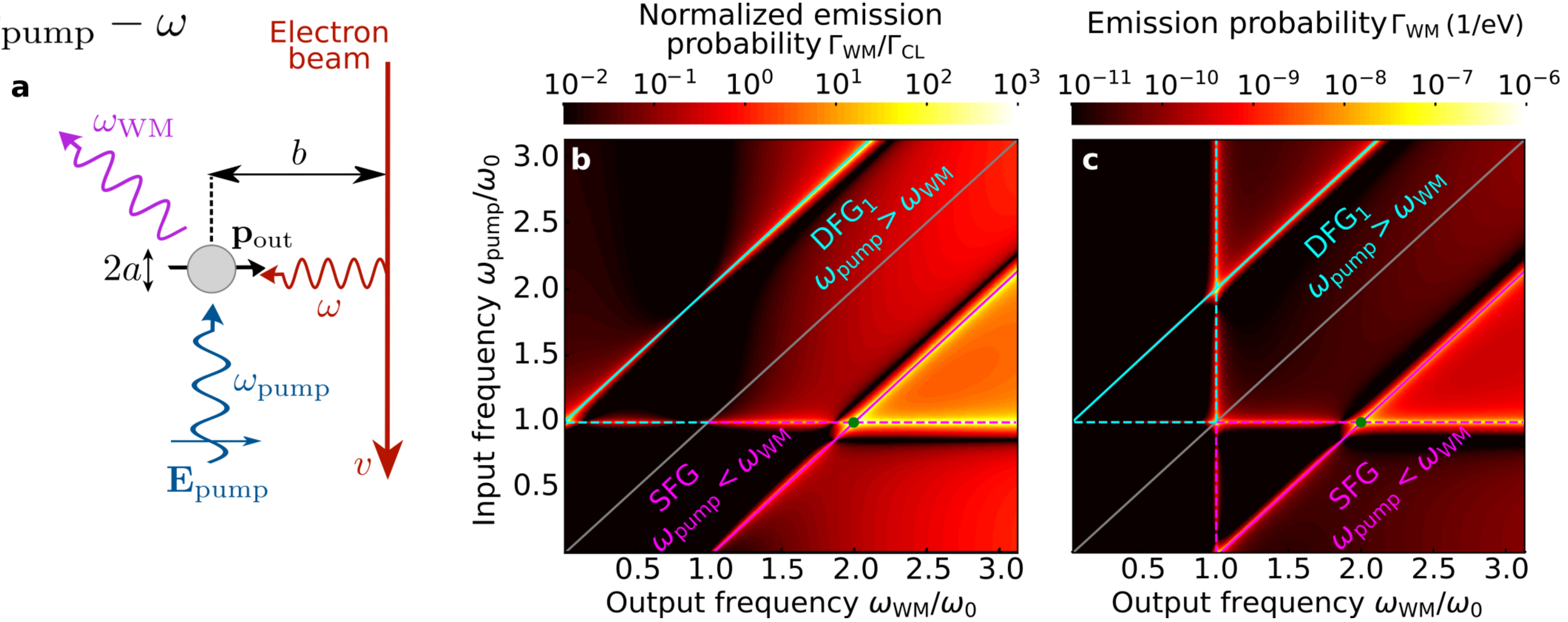
Emission rate of output photons:

$$\Gamma_{\mathrm{WM}}(\omega_{\mathrm{WM}}) \propto \frac{\omega_{\mathrm{WM}}^3\omega^2}{c^3 v^4 \gamma^2} K_1^2\left(\frac{\omega b}{v\gamma}\right) \frac{\left|\mathbf{E}_{\mathrm{pump}}\right|^2\left|\chi^{(2)}\right|^2}{\boxed{\left|[\epsilon(\omega_{\mathrm{pump}})+2][\epsilon(\omega_{\mathrm{WM}})+2][\epsilon(\omega)+2]\right|^2}} \quad \omega_0$$

Emission of photons enhanced at the resonant frequency

SFG: $\omega_{\mathrm{WM}} = \omega_{\mathrm{pump}} + \omega$

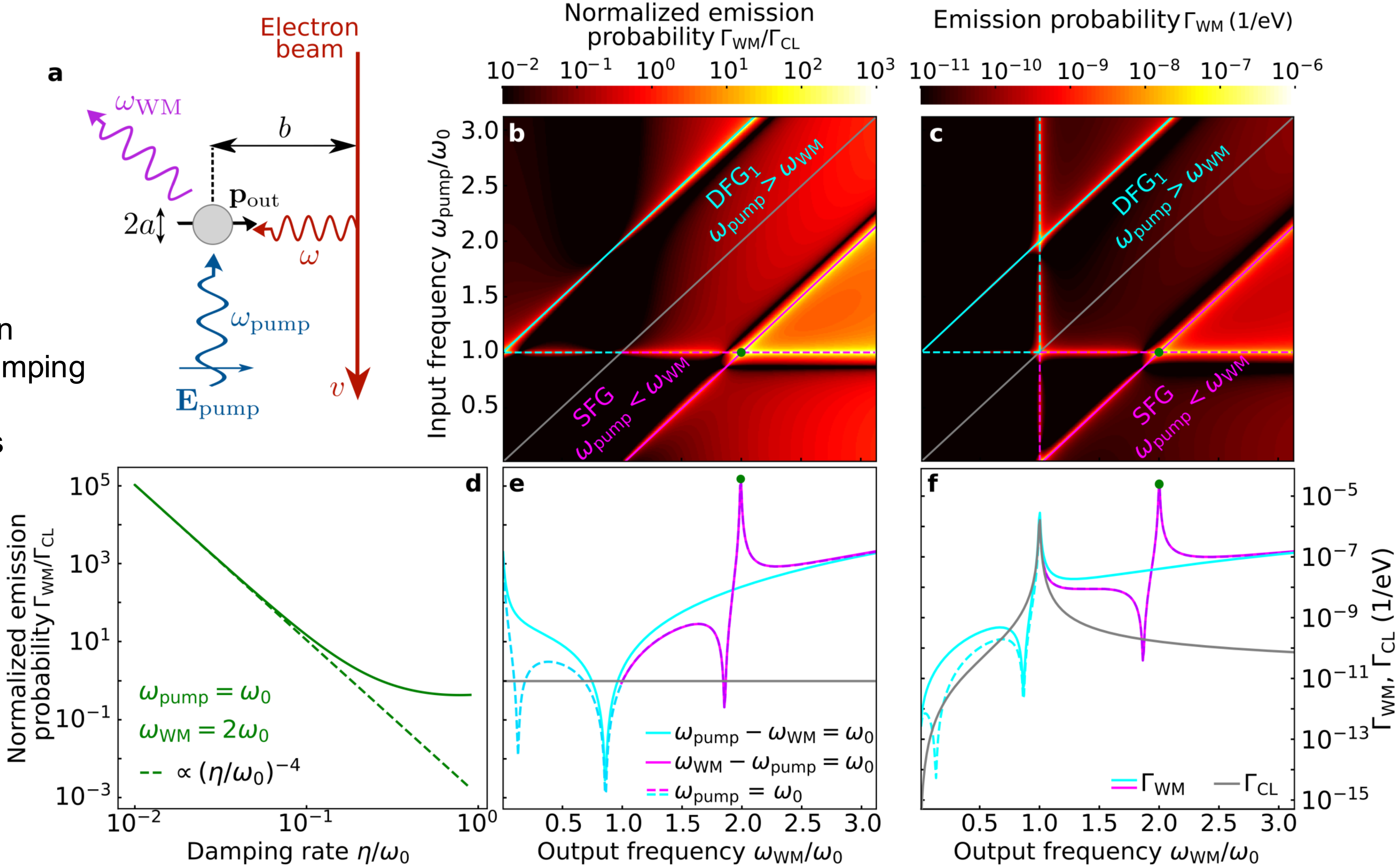
DFG$_1$: $\omega_{\mathrm{WM}} = \omega_{\mathrm{pump}} - \omega$



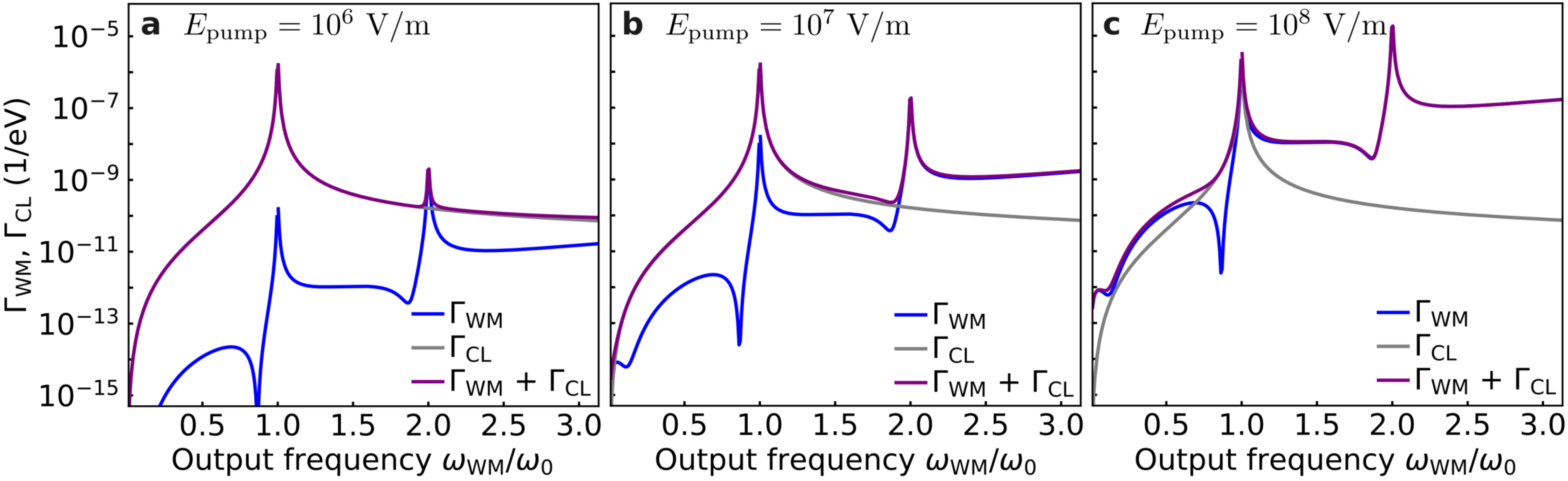
Emission rate of output photons for an electrostatic cavity

-Maximum photon emission rate: damping

-Universal results



How to isolate WMCL from regular CL



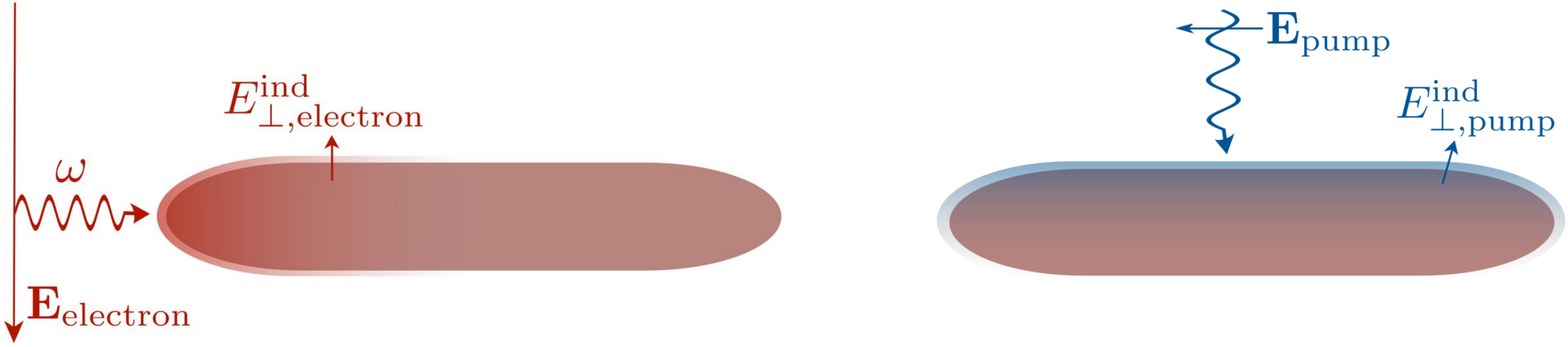
- CL background (grey) is pump-independent
- WM signal (blue) grows with pump field
- Total signal with laser on: WM + CL (purple) → what is measured
- Signal with laser off: CL → substract this signal

Emission rate of output photons for a large cavity

We extend the study to large cavities

Induced dipole moment:

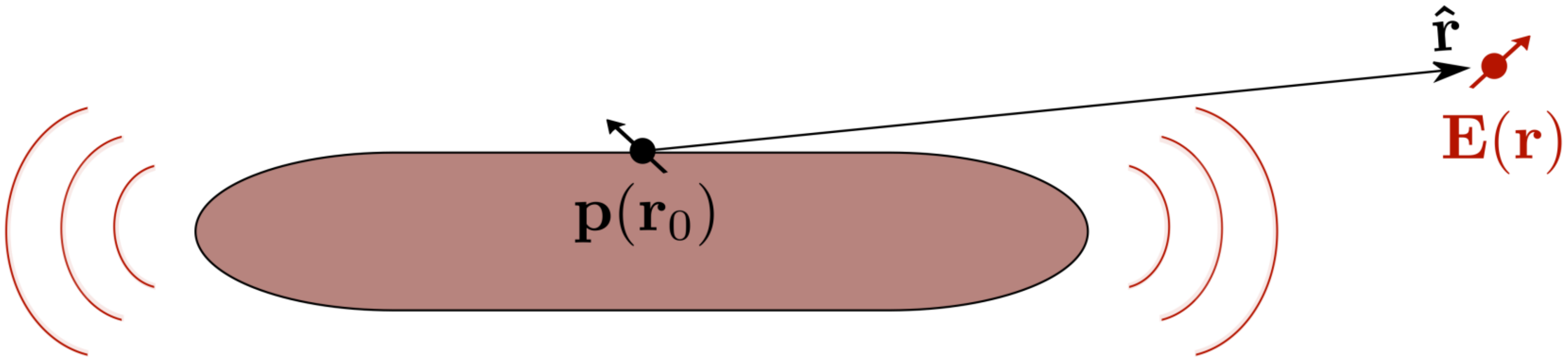
$$\mathbf{p}_{\mathrm{WM}}(\mathbf{r}_{\mathrm{s}},\omega_{\mathrm{WM}}) = \chi^{(2)}_{\perp\perp\perp}(\omega_{\mathrm{pump}},\omega,\omega_{\mathrm{WM}})E^{\mathrm{ind}}_{\perp,\mathrm{electron}}(\mathbf{r}_{\mathrm{s}},\omega)\,E^{\mathrm{ind}}_{\perp,\mathrm{light}}(\mathbf{r}_{\mathrm{s}},\omega_{\mathrm{pump}})\hat{\mathbf{n}}_{\mathrm{s}}$$



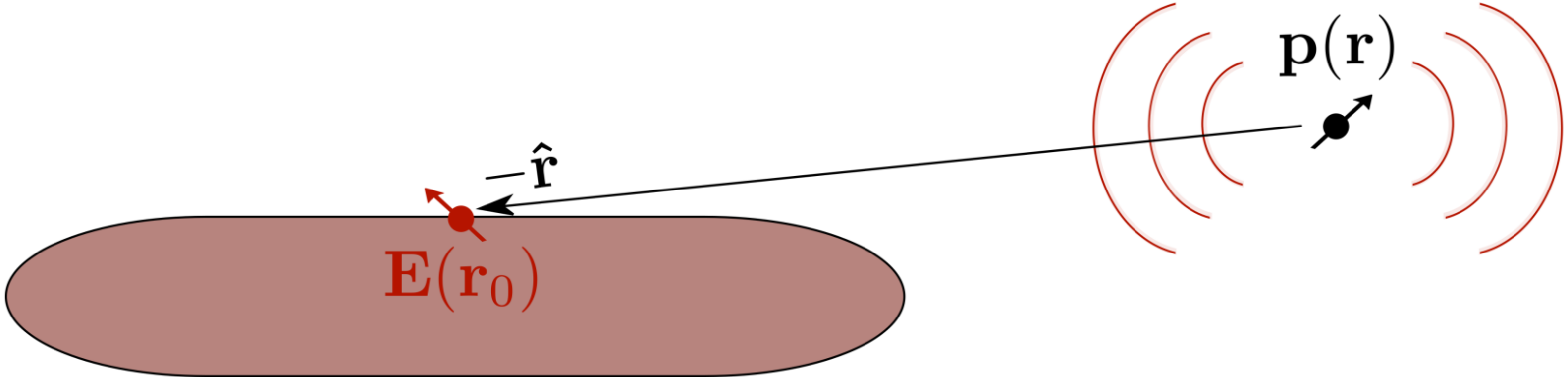
Emission rate of output photons for a large cavity

Reciprocity theorem: $\mathcal{G}(\mathbf{r}, \mathbf{r}_0, \omega) = \mathcal{G}^{\mathrm{T}}(\mathbf{r}_0, \mathbf{r}, \omega)$

A dipole radiating a field



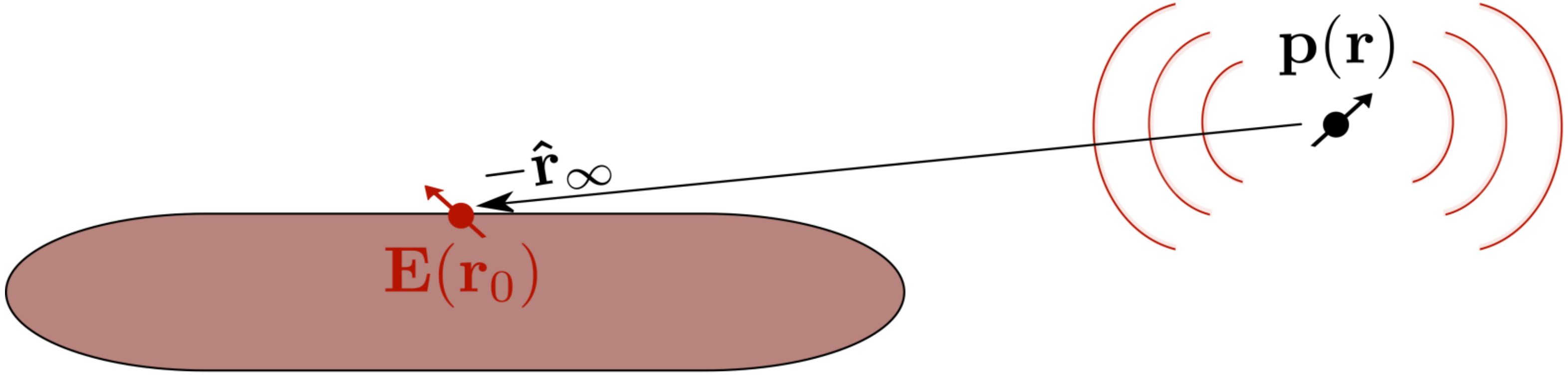
Having a "reciprocal" dipole



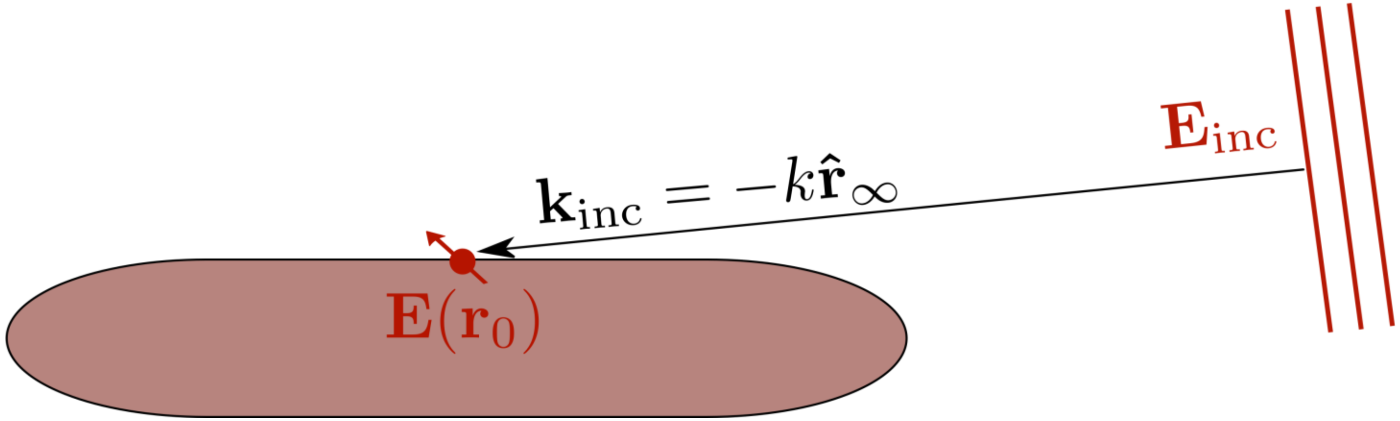
Emission rate of output photons for a large cavity

Equivalent plane-wave illumination:

The far-field "reciprocal" dipole



Plane-wave travelling



Emission rate of output photons for a large cavity

$$\mathbf{p}_{\mathrm{WM}}(\mathbf{r}_{\mathrm{s}},\omega_{\mathrm{WM}}) = \chi^{(2)}_{\perp\perp\perp}(\omega_{\mathrm{pump}},\omega,\omega_{\mathrm{WM}})E^{\mathrm{ind}}_{\perp,\mathrm{electron}}(\mathbf{r}_{\mathrm{s}},\omega)\,E^{\mathrm{ind}}_{\perp,\mathrm{light}}(\mathbf{r}_{\mathrm{s}},\omega_{\mathrm{pump}})\hat{\mathbf{n}}_{\mathrm{s}}$$

Induced dipole moment leads to a far-field distribution:

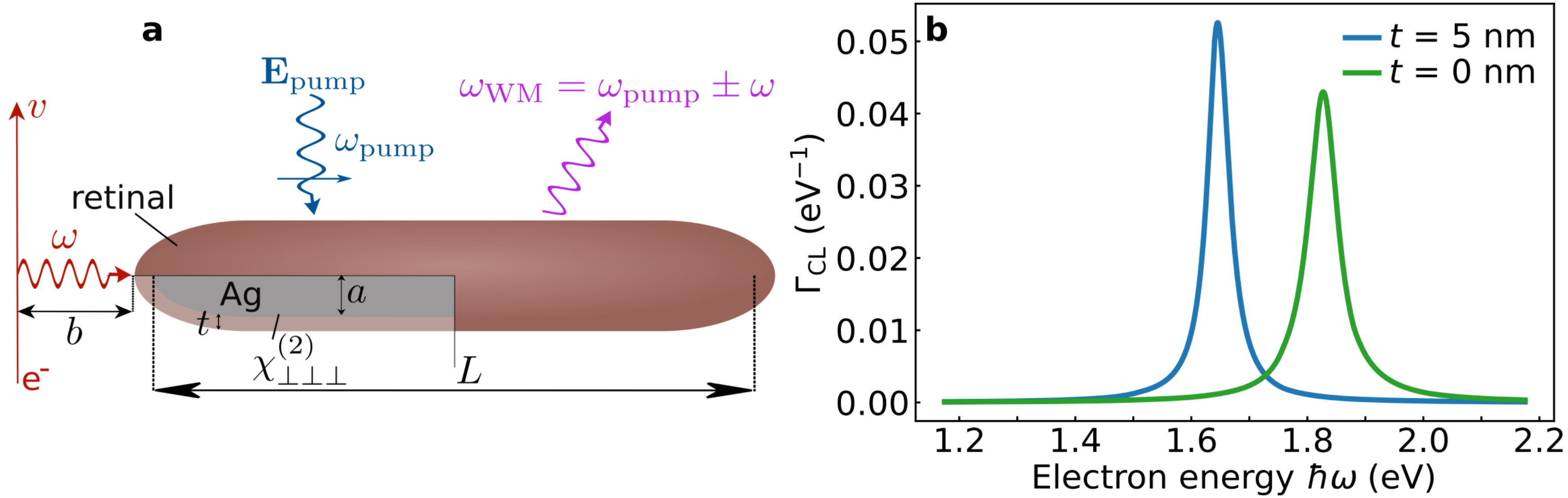
$$\mathbf{f}_{\mathrm{WM}}(\hat{\mathbf{r}},\omega_{\mathrm{WM}}) = k^2_{\mathrm{WM}}\sum_{\sigma=\mathrm{s,p}}\int_S d^2\mathbf{r}_{\mathrm{s}}\,\underset{\text{Equivalent plane-wave illumination}}{\boxed{\left[\frac{\mathbf{E}_\sigma(\mathbf{r}_{\mathrm{s}},\hat{\mathbf{r}},\omega_{\mathrm{WM}})}{E_0}\right]}}\,\underset{\text{Electron and light induced fields}}{\boxed{\mathbf{p}_{\mathrm{WM}}(\mathbf{r}_{\mathrm{s}},\omega_{\mathrm{WM}})}}$$

Wave-mixing photons emitted:

$$\Gamma_{\mathrm{WM}}(\omega_{\mathrm{WM}}) = \frac{c}{4\pi^2\hbar\omega_{\mathrm{WM}}}\int \mathrm{d}\Omega_{\hat{\mathbf{r}}}\,\left|\mathbf{f}_{\mathrm{WM}}(\hat{\mathbf{r}},\omega_{\mathrm{WM}})\right|^2$$

Emission rate of output photons for a large cavity

The cavity is a silver nanorod covered in retinal:

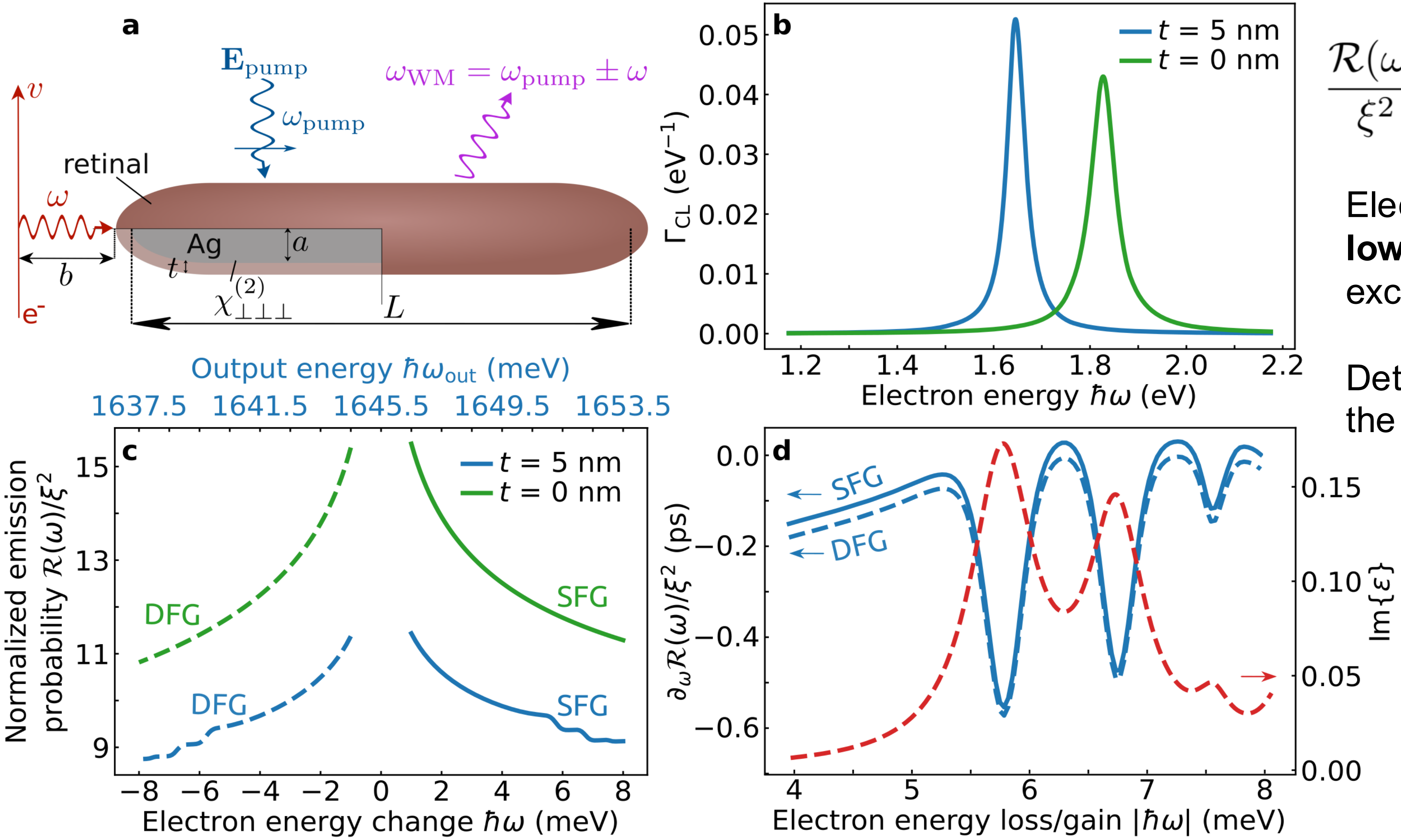


Silver nanorod act as a **plasmonic** antenna

Pump light fixed to the **visible** resonance of the nanorod

Emission rate of output photons for a large cavity

The cavity is a silver nanorod covered in retinal:

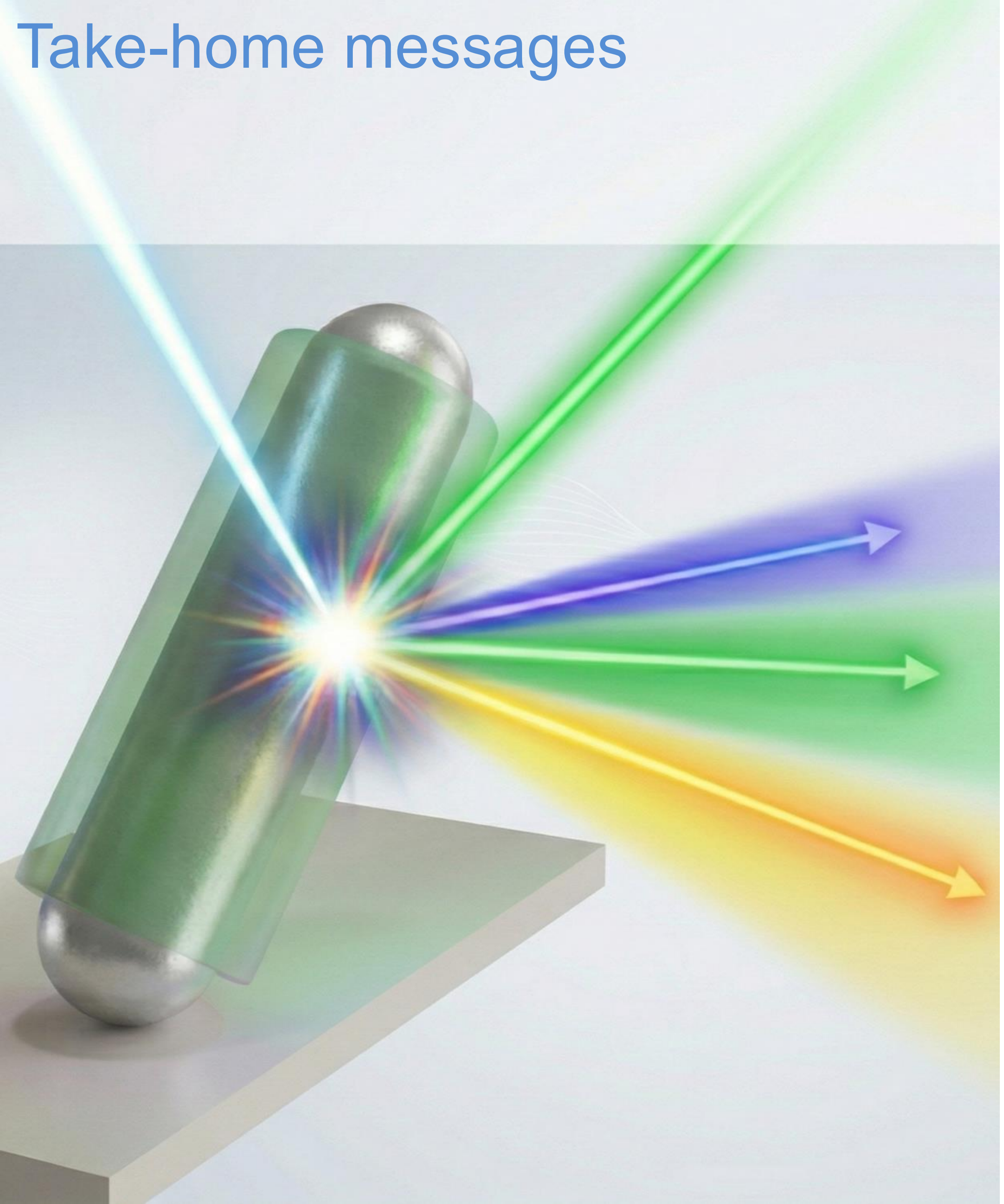


$$\frac{\mathcal{R}(\omega)}{\xi^2} \propto \frac{\Gamma_{\rm SFG/DFG}(\omega_{\rm WM})}{|\chi^{(2)}E_0|^2\Gamma_{\rm CL}(\omega_{\rm WM})}$$

Electron: provides broadband **low-frequency** drive that excite the retinal vibrations

Detect visible photons from the energy shift

Take-home messages



- WMCL = electron near field + optical pump mixed in a nonlinear cavity
- Low-frequency resonances become visible-light shifts
- WMCL aims to combine nanoscale localization from electrons with easy visible-light illumination and detection

Thank you!

Prelat, L., Dias, E.J.C. & García de Abajo, F.J. Wave-mixing cathodoluminescence microscopy of low-frequency excitations. *Nat Commun* **16**, 11551 (2025)

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MINISTERIO DE CIENCIA E INNOVACIÓN

EBEAM
Electron Beams Enhancing Analytical Microscopy

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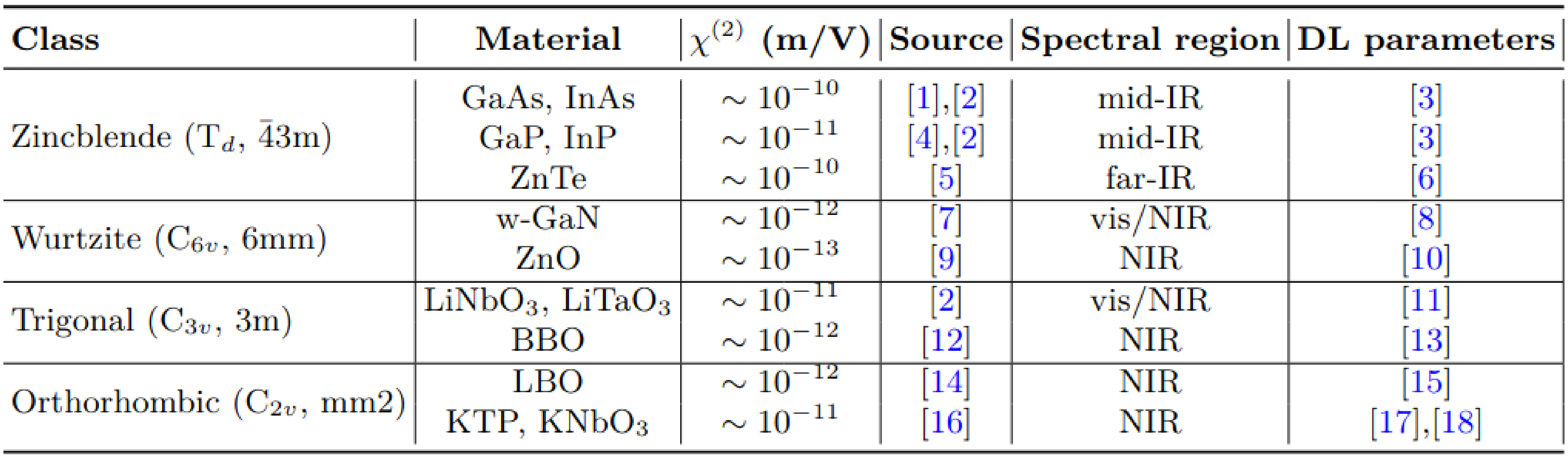
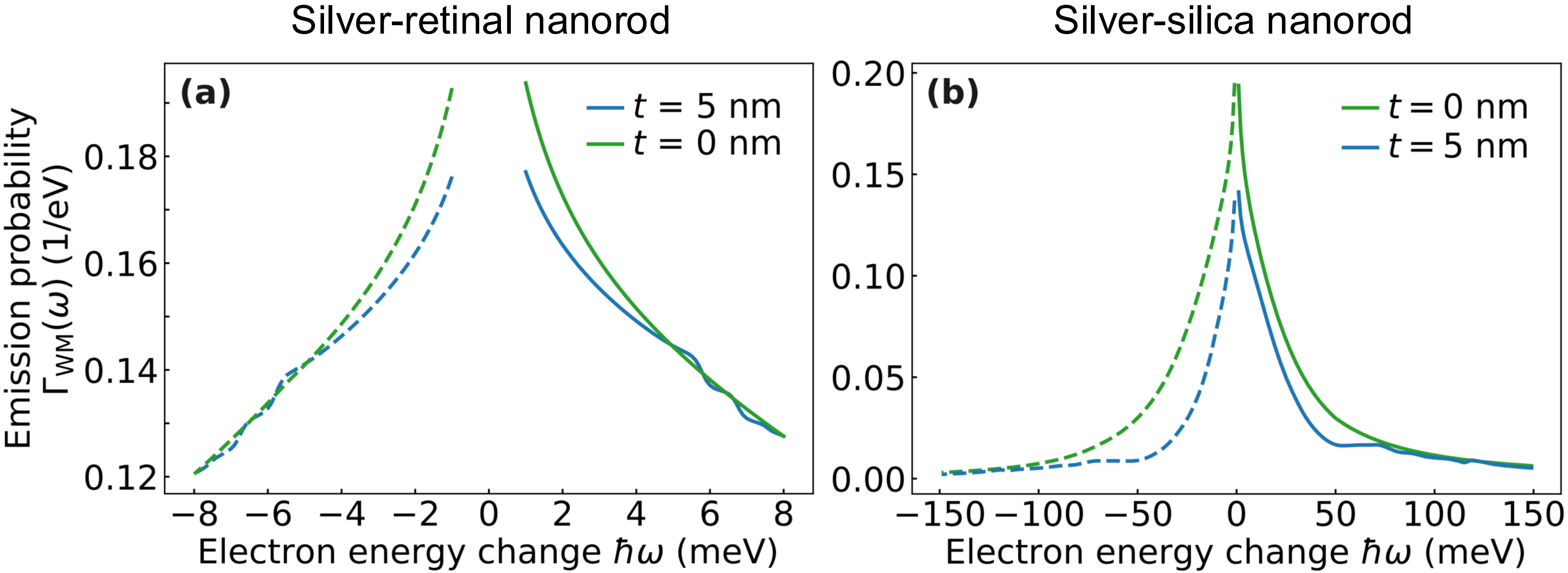
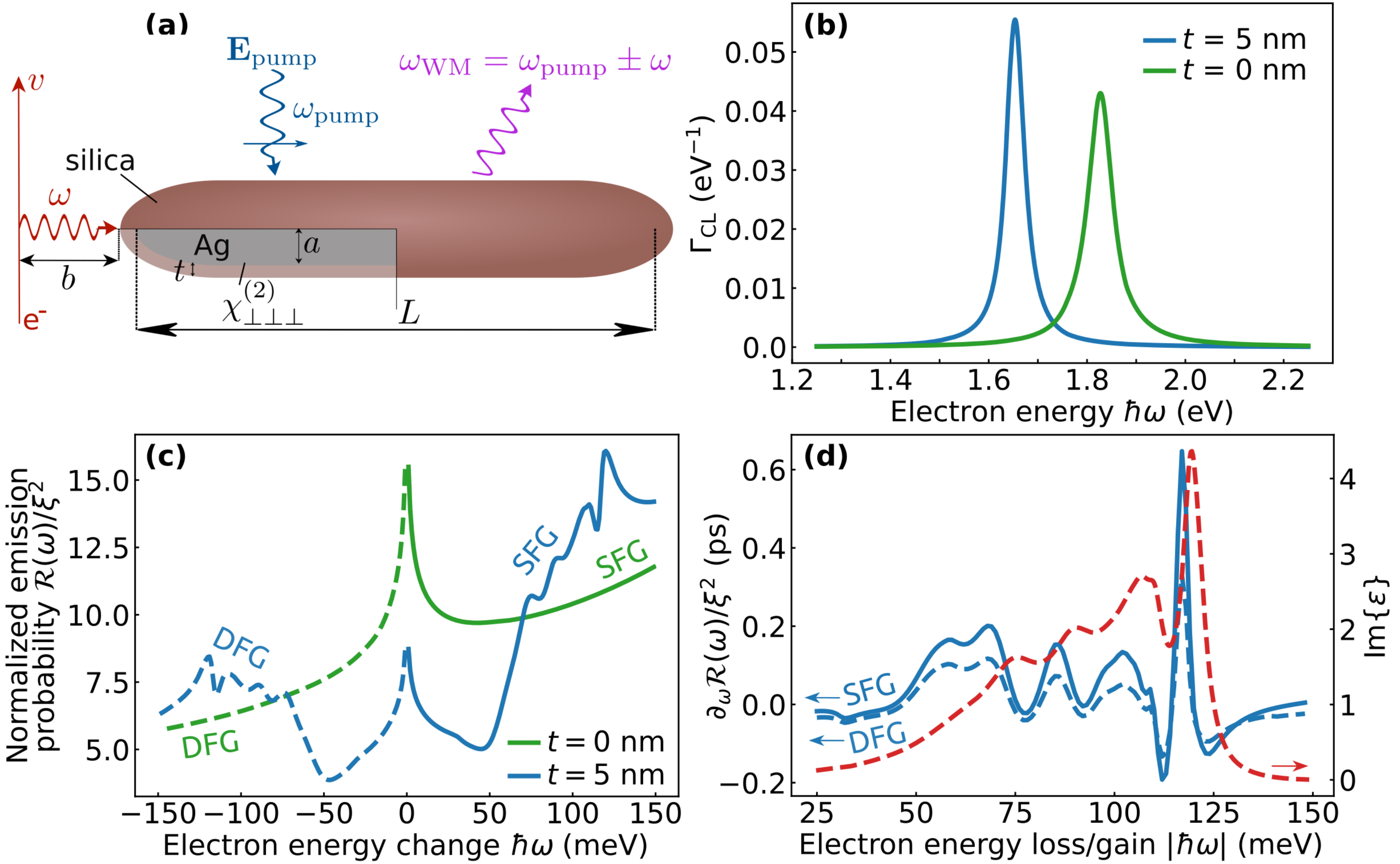
Representative bulk nonlinear susceptibility magnitudes for non-centrosymmetric materials



Class	**Material**	$\chi^{(2)}$ **(m/V)**	**Source**	**Spectral region**	**DL parameters**
Zincblende (T_d, $\bar{4}3m$)	GaAs, InAs	$\sim 10^{-10}$	[1],[2]	mid-IR	[3]
	GaP, InP	$\sim 10^{-11}$	[4],[2]	mid-IR	[3]
	ZnTe	$\sim 10^{-10}$	[5]	far-IR	[6]
Wurtzite (C_{6v}, 6mm)	w-GaN	$\sim 10^{-12}$	[7]	vis/NIR	[8]
	ZnO	$\sim 10^{-13}$	[9]	NIR	[10]
Trigonal (C_{3v}, 3m)	$LiNbO_3$, $LiTaO_3$	$\sim 10^{-11}$	[2]	vis/NIR	[11]
	BBO	$\sim 10^{-12}$	[12]	NIR	[13]
Orthorhombic (C_{2v}, mm2)	LBO	$\sim 10^{-12}$	[14]	NIR	[15]
	KTP, $KNbO_3$	$\sim 10^{-11}$	[16]	NIR	[17],[18]

Prelat, L., Dias, E.J.C. & García de Abajo, F.J. Wave-mixing cathodoluminescence microscopy of low-frequency excitations. *Nat Commun* **16**, 11551 (2025). https://doi.org/10.1038/s41467-025-67288-1


Silver-retinal nanorod
Silver-silica nanorod
(a)
t = 5 nm
t = 0 nm
Emission probability
$\Gamma_{\mathrm{WM}}(\omega)$ (1/eV)
0.12
0.14
0.16
0.18
−8
−6
−4
−2
0
2
4
6
8
Electron energy change $\hbar\omega$ (meV)
(b)
$t = 0$ nm
$t = 5$ nm
0.00
0.05
0.10
0.15
0.20
−150
−100
−50
0
50
100
150
Electron energy change $\hbar\omega$ (meV)


(a)
$\mathbf{E}_{\mathrm{pump}}$
ω_{pump}
$\omega_{\mathrm{WM}} = \omega_{\mathrm{pump}} \pm \omega$
v
silica
ω
Ag
a
t
b
$\chi^{(2)}_{\perp\perp\perp}$
L
e-
(b)
t = 5 nm
t = 0 nm
Γ_{CL} (eV^{-1})
0.05
0.04
0.03
0.02
0.01
0.0
1.2
1.4
1.6
1.8
2.0
2.2
Electron energy $\hbar\omega$ (eV)
(c)
Normalized emission probability $\mathcal{R}(\omega)/\xi^2$
15.0
12.5
10.0
7.5
5.0
DFG
DFG
SFG
SFG
$t = 0$ nm
$t = 5$ nm
−150
−100
−50
0
50
100
150
Electron energy change $\hbar\omega$ (meV)
(d)
$\partial_\omega \mathcal{R}(\omega)/\xi^2$ (ps)
0.6
0.4
0.2
0.0
−0.2
SFG
DFG
Im{ε}
4
3
2
1
0
25
50
75
100
125
150
Electron energy loss/gain $|\hbar\omega|$ (meV)

Emission rate of output photons for a large cavity

Induced dipole moment leads to a far-field distribution:

$$\mathbf{f}_{\mathrm{out}}(\hat{\mathbf{r}}) = k_{\mathrm{out}}^2 (1 - \hat{\mathbf{r}} \otimes \hat{\mathbf{r}}) \int_S \mathrm{d}^2\mathbf{r}_{\mathrm{s}}\, \mathrm{e}^{-\mathrm{i}k_{\mathrm{out}}\hat{\mathbf{r}}\cdot\mathbf{r}_{\mathrm{s}}} \mathbf{p}_{\mathrm{out}}(\mathbf{r}_{\mathrm{s}})$$

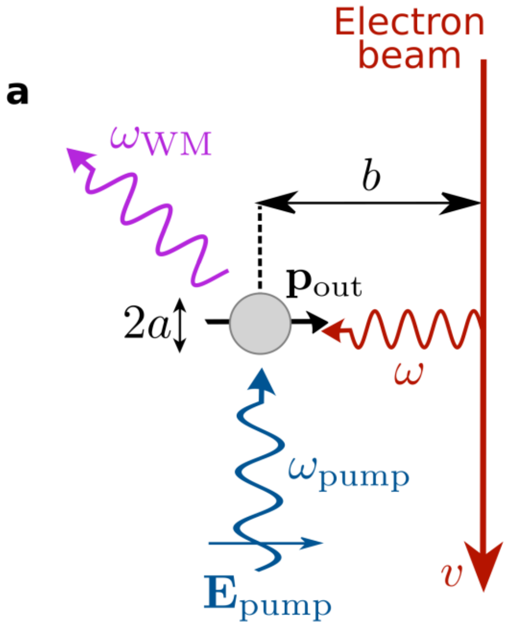
Far-field using the reciprocity theorem:

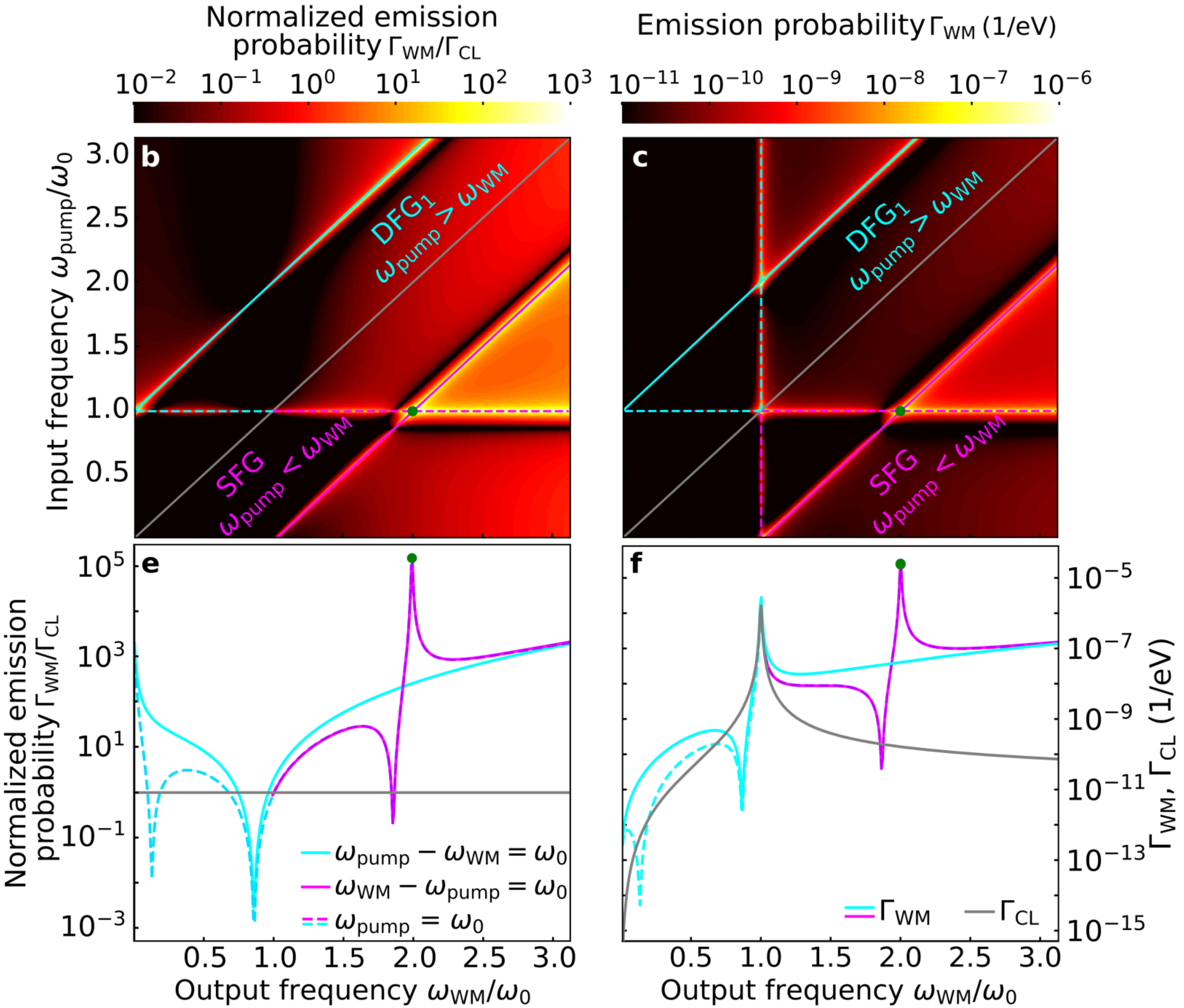
$$\mathbf{f}_{\mathrm{out}}(\hat{\mathbf{r}}) = k_{\mathrm{out}}^2 \sum_{j=\mathrm{s,p}} \int_S \mathrm{d}^2\mathbf{r}_{\mathrm{s}} \left(\frac{\mathbf{E}_j(\mathbf{r}_{\mathrm{s}}, \hat{\mathbf{r}}) \cdot \mathbf{p}_{\mathrm{out}}(\mathbf{r}_{\mathrm{s}})}{E_0} \right)$$

Output photons emitted:

$$\Gamma_{\mathrm{out}}(\omega) = \frac{1}{\hbar\omega_{\mathrm{out}}} \frac{c}{2\pi} \int \mathrm{d}\Omega\, |\mathbf{f}_{\mathrm{out}}(\hat{\mathbf{r}})|^2$$

Emission rate of output photons for an electrostatic cavity





SFG: $\omega_{\rm WM} = \omega_{\rm pump} + \omega$

DFG$_1$: $\omega_{\rm WM} = \omega_{\rm pump} - \omega$

The emission of photons is enhanced for the resonant frequency ω_0