

Supplementary Material to the article “Second-Harmonic Generation under Irradiation of Superconducting Films with Structured Light”

The Supplementary Material presents the details omitted from the main text: the linearization of the time-dependent Ginzburg–Landau equation, the boundary conditions for the amplitude modulation Δ_1 and the charge-imbalance potential $\tilde{\phi}_\omega$, the transition to the thin-film limit, and the derivation of the radiation outcoupling coefficients at the frequency 2ω . The basic geometry is a film occupying the region $0 < z < d$, illuminated from the side $z < 0$. All quantities carrying the index ω are complex amplitudes with the time dependence $\exp(-i\omega t)$.

1. *Linearized Ginzburg–Landau equation and boundary conditions.* The initial time-dependent Ginzburg–Landau equation has the form

$$-\Gamma(\hbar\partial_t - 2ie\phi)\Delta = -aT_c\epsilon\Delta + b|\Delta|^2\Delta + aT_c\xi_0^2\left(-i\nabla + \frac{2\pi}{\Phi_0}\mathbf{A}\right)^2\Delta. \quad (\text{S1})$$

Here $\epsilon = (T_c - T)/T_c$, $\Gamma = \pi a/8 + i\gamma$, and $\nu = 8\gamma/(\pi a)$ characterizes the electron–hole asymmetry. We represent the order parameter in the form

$$\Delta = (\Delta_0 + \Delta_1)e^{i\chi}, \quad \Delta_0^2 = \frac{aT_c\epsilon}{b}, \quad (\text{S2})$$

and introduce the gauge-invariant charge-imbalance potential

$$\tilde{\phi} = \phi - \mu_p, \quad \mu_p = \frac{\hbar}{2e}\partial_t\chi. \quad (\text{S3})$$

The real part of Eq. (S1), linearized with respect to Δ_1 , gives

$$-\tau\partial_t\Delta_1 - \tau\nu\Delta_0\frac{2e}{\hbar}\tilde{\phi} = 2\Delta_1 - \xi^2\nabla^2\Delta_1, \quad (\text{S4})$$

where

$$\xi^2 = \frac{\xi_0^2}{\epsilon}, \quad \tau = \frac{\pi\hbar}{8T_c\epsilon}. \quad (\text{S5})$$

For the first harmonic proportional to $\exp(-i\omega t)$, we obtain

$$-\nabla^2\Delta_1 + q_1^2\Delta_1 = -\frac{2e\tau\nu\Delta_0}{\hbar\xi^2}\tilde{\phi}_\omega, \quad q_1^2 = \frac{2 - i\omega\tau}{\xi^2}. \quad (\text{S6})$$

The imaginary part of Eq. (S1), together with the continuity equation, leads to

$$\nabla^2\tilde{\phi}_\omega = q_2^2\tilde{\phi}_\omega, \quad q_2^2 = l_E^{-2} - \frac{i\omega\tau}{\xi^2} = \frac{\lambda^2}{l_E^2\lambda_{\text{eff}}^2}, \quad (\text{S7})$$

where

$$l_E^2 = \frac{\hbar\sigma_n}{\pi a e^2 \Delta_0^2}, \quad \lambda^2 = \frac{\hbar^2 c^2}{32\pi a T_c \xi_0^2 e^2 \Delta_0^2}, \quad (\text{S8})$$

$$\lambda_{\text{eff}}^{-2} = \lambda^{-2} - \frac{4\pi i \sigma_n \omega}{c^2} = \lambda^{-2} \left(1 - \frac{i\omega\tau l_E^2}{\xi^2}\right). \quad (\text{S9})$$

For a surface that does not suppress superconductivity, the natural boundary condition for the order parameter is

$$\mathbf{n} \cdot \left(-i\nabla + \frac{2\pi}{\Phi_0}\mathbf{A}\right)\Delta \Big|_{\text{surf}} = 0. \quad (\text{S10})$$

Substitution of Eq. (S2) and the separate vanishing of the amplitude and phase parts give, to linear order in $\mathbf{A}$,

$$\mathbf{n} \cdot \nabla\Delta_1 \Big|_{\text{surf}} = 0, \quad \mathbf{n} \cdot \mathbf{v}_s \Big|_{\text{surf}} = 0. \quad (\text{S11})$$

For the charge-imbalance potential, we use the boundary condition

$$\partial_n \tilde{\phi}_\omega \Big|_{\text{surf}} = -E_n^{(i)} \Big|_{\text{surf}}, \quad (\text{S12})$$

where $\mathbf{E}^{(i)}$ is the field inside the superconductor beyond the thin Thomas–Fermi screening layer. For a film occupying the region $0 < z < d$, this condition takes the form

$$\partial_z \tilde{\phi}_\omega \Big|_{z=0} = -E_z^{(i)}(0), \quad \partial_z \tilde{\phi}_\omega \Big|_{z=d} = -E_z^{(i)}(d). \quad (\text{S13})$$

Within the assumption of almost complete reflection at the fundamental frequency used in this work, the field at the surface $z = d$ is small, and therefore we set $E_z^{(i)}(d) \simeq 0$ below. For an individual plane-wave component of the angular spectrum with incidence angle θ and amplitude α of the P -polarized component, the normal field at the illuminated surface is

$$E_z^{(i)}(0) = \frac{i\alpha\omega \sin\theta}{2\pi\sigma_n} B_0. \quad (\text{S14})$$

The spatial factor of this component is omitted in Eq. (S14); for a Laguerre–Gaussian beam, the formula is applied to each component of its angular spectrum.

2. *Amplitude modulation and the thin-film source.* For brevity, we introduce

$$h_j(z) = \frac{\cosh q_j(d-z)}{q_j \sinh q_j d}, \quad j = 1, 2. \quad (\text{S15})$$

These functions satisfy the conditions $h_j'(0) = -1$ and $h_j'(d) = 0$. Therefore, the solution of Eq. (S7) with the boundary conditions (S13) is

$$\tilde{\phi}_\omega(z) = E_z^{(i)}(0)h_2(z). \quad (\text{S16})$$

In particular, the value of the potential at the illuminated surface is

$$\begin{aligned} \tilde{\phi}_\omega(0) &= \frac{E_z^{(i)}(0)}{q_2} \coth(q_2 d) \\ &= \frac{i\alpha\omega \sin\theta}{2\pi\sigma_n q_2} \coth(q_2 d) B_0. \end{aligned} \quad (\text{S17})$$

In the half-space limit $d \rightarrow \infty$, this gives $\tilde{\phi}_\omega(0) = E_z^{(i)}(0)/q_2$.

The solution of Eq. (S6) satisfying the conditions $\partial_z \Delta_1 = 0$ at $z = 0$, d can be conveniently written directly in terms of the functions (S15):

$$\Delta_1(z) = \frac{2e\tau\nu\Delta_0}{\hbar\xi^2} \frac{E_z^{(i)}(0)}{q_1^2 - q_2^2} [h_1(z) - h_2(z)]. \quad (\text{S18})$$

Equations (S16) and (S18) are required only when the profile across the film thickness is substantially nonuniform. In the main text, we use the thin-film limit

$$|q_1|d \ll 1, \quad |q_2|d \ll 1, \quad \frac{d}{|\lambda_{\text{eff}}|} \ll 1. \quad (\text{S19})$$

The potential and the amplitude correction are then uniform across the film thickness with relative accuracy of order d^2 :

$$\tilde{\phi}_\omega(z) = \frac{E_z^{(i)}(0)}{q_2^2 d} [1 + \mathcal{O}((q_2 d)^2)], \quad (\text{S20})$$

$$\Delta_1(z) = \Delta_\omega [1 + \mathcal{O}((q_1 d)^2 + (q_2 d)^2)],$$

$$\Delta_\omega = -\frac{2e\tau\nu\Delta_0}{\hbar\xi^2} \frac{E_z^{(i)}(0)}{dq_1^2 dq_2^2}. \quad (\text{S21})$$

Under the third condition in Eq. (S19), the linear superfluid velocity is also practically independent of z :

$$\mathbf{v}_{s,\omega}(z) = \mathbf{v}_{s,\omega} [1 + \mathcal{O}(d^2/\lambda_{\text{eff}}^2)], \quad (\text{S22})$$

where $\mathbf{v}_{s,\omega}$ is the main text. Therefore, both nonlinear sources can be immediately replaced by their mean volume densities,

$$\begin{aligned} \bar{\mathbf{j}}_{\text{eh},2\omega} &= -2e\Delta_0\Delta_\omega\mathbf{v}_{s,\omega}, \\ \bar{\mathbf{j}}_{\text{D},2\omega} &= \frac{e\Delta_0^2}{2v_c} [(\mathbf{v}_{s,\omega} \cdot \mathbf{v}_{s,\omega})\mathbf{u}_0 + 2(\mathbf{u}_0 \cdot \mathbf{v}_{s,\omega})\mathbf{v}_{s,\omega}]. \end{aligned} \quad (\text{S23})$$

Thus, in the working limit of the main text, a separate normalized profile function of the superfluid velocity is not required: the source is taken to be constant for $0 < z < d$. If any of the conditions in Eq. (S19) is violated, the actual dependence $\mathbf{j}_{2\omega}(q, z)$, constructed from Eqs. (S16), (S18), and the solution of the linear electrodynamic problem for $\mathbf{v}_{s,\omega}(z)$, should be used in the formulas of the next section.

3. Radiation outcoupling from the film. After the Fourier-Bessel transformation in the film plane, each transverse circular component of the field at the frequency 2ω satisfies the equation

$$(\partial_z^2 - \kappa_2^2) E(q, z) = -\frac{8\pi i \omega}{c^2} j(q, z), \quad (\text{S24})$$

inside the film, where

$$\lambda_2^{-2} = \lambda^{-2} - \frac{8\pi i \omega \sigma_n}{c^2}, \quad \kappa_2(q) = \sqrt{\lambda_2^{-2} + q^2}. \quad (\text{S25})$$

In vacuum,

$$K_2(q) = \sqrt{k_2^2 - q^2}, \quad k_2 = \frac{2\omega}{c}. \quad (\text{S26})$$

The sign of the expression under the square root in Eq. (S26) simultaneously specifies outgoing waves for $q < k_2$ and evanescent fields for $q > k_2$.

Let

$$j(q, z) = j(q, 0)f(z), \quad f(0) = 1, \quad (\text{S27})$$

and define the function $g(z)$ by

$$E(q, z) = -\frac{8\pi i \omega}{c^2} j(q, 0)g(z). \quad (\text{S28})$$

Then

$$g'' - \kappa_2^2 g = f(z). \quad (\text{S29})$$

Outside the film, we retain only the waves propagating away from the sample:

$$\begin{aligned} E^<(q, z) &= -\frac{8\pi i \omega}{c^2} j(q, 0)T_-(q)e^{-iK_2 z}, \quad z < 0, \\ E^>(q, z) &= -\frac{8\pi i \omega}{c^2} j(q, 0)T_+(q)e^{iK_2(z-d)}, \quad z > d. \end{aligned} \quad (\text{S30})$$

Continuity of the tangential electric- and magnetic-field components is equivalent to the two radiation boundary conditions

$$\begin{aligned} g'(0) &= -iK_2 g(0), & T_- &= g(0), \\ g'(d) &= iK_2 g(d), & T_+ &= g(d). \end{aligned} \quad (\text{S31})$$

The solution satisfying the upper boundary condition in Eq. (S31) can be written as

$$\begin{aligned} g(z) &= T_- \left[\cosh \kappa_2 z - \frac{iK_2}{\kappa_2} \sinh \kappa_2 z \right] \\ &\quad + \int_0^z \frac{\sinh \kappa_2(z-z')}{\kappa_2} f(z') dz'. \end{aligned} \quad (\text{S32})$$

Substitution of the bottom boundary condition from Eq. (S31) gives the radiation coefficient toward the region $z < 0$:

$$T_-[f] = -\frac{\int_0^d W_-(z)f(z) dz}{\mathcal{D}(q)}, \quad (\text{S33})$$

where

$$W_-(z) = \cosh \kappa_2(d-z) - \frac{iK_2}{\kappa_2} \sinh \kappa_2(d-z). \quad (\text{S34})$$

Similarly, for radiation toward the region $z > d$,

$$T_+[f] = -\frac{\int_0^d W_+(z)f(z) dz}{\mathcal{D}(q)}, \quad (\text{S35})$$

$$W_+(z) = \cosh \kappa_2 z - \frac{iK_2}{\kappa_2} \sinh \kappa_2 z. \quad (\text{S36})$$

The common denominator is

$$\mathcal{D}(q) = \frac{\kappa_2^2 - K_2^2}{\kappa_2} \sinh(\kappa_2 d) - 2iK_2 \cosh(\kappa_2 d). \quad (\text{S37})$$

Equations (S33) and (S35) have a simple interpretation: the kernels $W_\pm(z)$ describe propagation of the field from a source layer at depth z to the corresponding surface, whereas the denominator $\mathcal{D}$ accounts for multiple screening inside the film. If the profile is symmetric, $f(z) = f(d - z)$, then $T_- = T_+$. A nonsymmetric profile, for example one produced by charge imbalance excited predominantly near the surface $z = 0$, generally gives $T_- \neq T_+$.

In the thin-film regime of Sec. 2, the source is uniform, $f(z) = 1$. Then

$$\begin{aligned} T_- &= T_+ \equiv T_f, \\ T_f &= -\frac{\kappa_2 \sinh(\kappa_2 d) + iK_2 [1 - \cosh(\kappa_2 d)]}{\kappa_2^2 \mathcal{D}(q)}. \end{aligned} \quad (\text{S38})$$

For $|\kappa_2|d \ll 1$ and $|K_2|d \ll 1$, Eq. (S38) first gives the more general thin-film expression

$$T_f(q) \simeq -\frac{1}{\kappa_2^2(q) - K_2^2(q) - 2iK_2(q)/d}. \quad (\text{S39})$$

For paraxial radiation, when $q|\lambda_2| \ll 1$ and $k_2|\lambda_2| \ll 1$, we have $\kappa_2^2 - K_2^2 \simeq \lambda_2^{-2}$, and Eq. (S39) takes the form used in the main text:

$$T_f(q) \simeq -\frac{\lambda_2^2}{1 - 2iK_2(q)\lambda_2^2/d}. \quad (\text{S40})$$

If, in addition, $2|K_2||\lambda_2|^2/d \ll 1$, the screening becomes local:

$$T_f \simeq -\lambda_2^2. \quad (\text{S41})$$

The coefficients $T_\pm$ in Eq. (S30) multiply the chosen reference *volume* current density $j(q, 0)$; their dimension is length squared. In the main text, the quantity $\mathcal{J}_{m',\pm}^{(\sigma)}(q)$ is understood as the transform of the mean nonlinear volume current density across the film thickness. If the sheet current integrated over the film thickness is used instead,

$$J(q) = \int_0^d j(q, z) dz = j(q, 0) \int_0^d f(z) dz, \quad (\text{S42})$$

the field should be written using the coefficient

$$\begin{aligned} \Theta_\pm[f] &= \frac{T_\pm[f]}{\int_0^d f(z) dz}, \\ j(q, 0)T_\pm[f] &= J(q)\Theta_\pm[f]. \end{aligned} \quad (\text{S43})$$

For a uniform source, $\Theta_f = T_f/d$.

Thus, the reduced formulas of the main text are applicable when the following conditions are simultaneously satisfied:

$$\begin{aligned} |q_1|d &\ll 1, & |q_2|d &\ll 1, & \frac{d}{|\lambda_{\text{eff}}|} &\ll 1, \\ |\kappa_2|d &\ll 1, & |K_2|d &\ll 1. \end{aligned} \quad (\text{S44})$$

The first three conditions allow the nonlinear source to be replaced by the uniform mean density (S23); the last two reduce the exact coefficient (S38) to the thin-film expression (S39). If any of these conditions is violated, the actual profile $j(q, z)$ should be retained and the exact functionals (S33) and (S35) should be used.

4. *Relation between the beam profile and the electromagnetic-field components.* In the main text, for compact notation, we introduce the transverse vector $\mathbf{Q}_\omega = \mathbf{n} \times \mathbf{B}_\omega$, where $\mathbf{n}$ is the normal to the film. The function $U_p^m(\rho)$ is the scalar radial envelope of the Laguerre–Gaussian mode:

$$U_p^m(\rho) = \left(\frac{\sqrt{2}\rho}{w_0} \right)^{|m|} L_p^{|m|} \left(\frac{2\rho^2}{w_0^2} \right) \exp \left(-\frac{\rho^2}{w_0^2} \right). \quad (\text{S45})$$

Let u_s and u_p denote the weights of the s - (TE-) and p - (TM-) polarizations, respectively. In the laboratory circular basis, $E_{\omega,\pm} = E_{\omega,x} \pm iE_{\omega,y}$ and $B_{\omega,\pm} = B_{\omega,x} \pm iB_{\omega,y}$, the transverse components of the incident paraxial beam take the form

$$E_{\omega,\pm} \simeq -B_0 i^m e^{i(m\pm 1)\varphi} \frac{u_p \mp iu_s}{k} \left(\partial_\rho \mp \frac{m}{\rho} \right) U_p^m(\rho), \quad (\text{S46})$$

$$B_{\omega,\pm} \simeq \mp iB_0 i^m e^{i(m\pm 1)\varphi} \frac{u_p \mp iu_s}{k} \left(\partial_\rho \mp \frac{m}{\rho} \right) U_p^m(\rho). \quad (\text{S47})$$

Here we used the relations $\mathbf{E}_{\omega,\parallel} \simeq -\mathbf{Q}_\omega$ and $B_{\omega,\pm} = \mp iQ_{\omega,\pm}$.

The longitudinal electric-field component follows from the transversality condition $\nabla \cdot \mathbf{E}_\omega = 0$. For propagation along $+z$, in the paraxial approximation, $E_{\omega,z} \simeq (i/k)\nabla_\parallel \cdot \mathbf{E}_{\omega,\parallel}$, which gives

$$\begin{aligned} E_{\omega,z} &\simeq -\frac{B_0 i^{m+1} u_p}{k^2} e^{im\varphi} \left(\partial_\rho^2 + \frac{1}{\rho} \partial_\rho - \frac{m^2}{\rho^2} \right) U_p^m(\rho) \\ &= \frac{4B_0 i^{m+1} u_p}{k^2 w_0^2} e^{im\varphi} \left(2p + |m| + 1 - \frac{\rho^2}{w_0^2} \right) U_p^m(\rho). \end{aligned} \quad (\text{S48})$$

Thus, $E_{\omega,z} \propto u_p$ and vanishes for pure s -polarization, as expected for the TE component. Compared with the transverse field, the longitudinal component is small in the paraxial parameter, $|E_{\omega,z}|/|E_{\omega,\parallel}| \sim (kw_0)^{-1}$.

In the electron-hole mechanism, the corresponding internal normal field $E_{\omega,z}^{(i)}$ enters the boundary condition $\partial_z \tilde{\phi}_\omega = -E_{\omega,z}^{(i)}$ and excites the charge-imbalance potential.

According to (S6)–(S21), the latter produces a correction Δ_1 to the amplitude of the order parameter and, consequently, to the superfluid density.