

Langdon effect in the realm of extreme ultraviolet source plasmasJ. Gonzalez ¹ and J. Sheil ^{1,2,*}¹*Advanced Research Center for Nanolithography, Science Park 106, 1098 XG Amsterdam, The Netherlands*²*Department of Physics and Astronomy, and LaserLaB, Vrije Universiteit Amsterdam, De Boelelaan 1081, 1081 HV Amsterdam, The Netherlands*

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Already some years ago, Langdon [*Phys. Rev. Lett.* **44**, 575 (1980)] proposed that inverse bremsstrahlung absorption in plasmas drives free electrons into non-Maxwellian distributions. Radiation-hydrodynamic simulations of plasma-based light sources, however, often (implicitly) assume Maxwellian-distributed electrons. In this paper, we quantify the effect of non-Maxwellian distributions on laser absorption and thermal conduction in laser-driven plasma light sources. For irradiation conditions $I_{\text{las}} \lambda_{\text{las}}^2 \in [10^{11}, 10^{13}] \text{ W cm}^{-2} \mu\text{m}^2$ and plasma parameters $Z_{\text{eff}}/T_e \in [0.2, 0.6] \text{ eV}^{-1}$, the electron distributions are predicted to be super-Gaussian of order $m \simeq 2.2\text{--}2.4$. As a result, laser absorption is calculated to be 10–20% lower than plasmas with Maxwellian-distributed electrons. A $\sim 10\text{--}30\%$ reduction of the Spitzer-Härm thermal conduction coefficient is also predicted.

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Radiation-hydrodynamic (rad-hydro) simulations play a crucial role in the design and interpretation of laser-produced plasma (LPP) experiments, such as those encountered in inertial confinement fusion (ICF) research [1–3], as well as laser-plasma radiation sources for nanolithography [4–11] and biological imaging [12–14]. The basic principle in these latter two applications is the same: a target (such as Mo [15], Sn [16], Xe [17], W [18], or Au [19]) is irradiated with moderate-intensity laser pulses ($I_{\text{las}} = 10^{10} - 10^{13} \text{ W cm}^{-2}$) to create plasmas that radiate intensely in application-specific wavelength regions. For nanolithography, this region is the $13.5 \pm 0.135 \text{ nm}$ band (the reflective wavelengths of molybdenum-silicon multilayer mirrors [20]) and for biological imaging interest lies in the water window region (2.3–4.4 nm) [12]. Identifying experimental conditions that maximize light source performance is a very challenging task, and it is here that predictive rad-hydro modeling can play a defining role.

While predictivity is an obvious criterion for any simulation capability, the many interacting components of rad-hydro simulations [21] (e.g., atomic kinetics [22,23], heat transport [24,25], and equation-of-state properties [26]), as well as their numerous possible implementations, complicates the testing and benchmarking of rad-hydro codes. An additional challenge arises when modeling laser absorption in a plasma. In 1980, Langdon [27] predicted that inverse bremsstrahlung absorption (IBA) drives free electrons into non-Maxwellian distributions so long as electron-electron collisions are not sufficiently frequent to establish a Maxwellian distribution.

The effect is inherently nonlinear; preferential heating of low-energy electrons results in a lower laser absorptivity than plasmas with a Maxwellian electron distribution function (EDF). The number of electrons in the high-energy tail is also suppressed compared to a Maxwellian EDF; see Fig. 1 of Matte *et al.* [28]. A correction factor is typically incorporated into rad-hydro simulations to account for the reduction in laser absorption [8].

Only in the recent experimental works of Turnbull *et al.* [29] and Milder *et al.* [30–32] were non-Maxwellian EDFs identified in high-temperature, ICF-relevant laser-driven plasmas. The aforementioned reduction in laser absorption was observed in the work of Turnbull *et al.* [33], where a subsequent analysis [34] revealed that accounting for (i) the super-Gaussian nature of the EDFs in deriving the Coulomb logarithm and (ii) a correction for ion screening were necessary to bring calculations into agreement with measurements. An impression of the cumulative impact of their findings for hohlraum simulations is presented in Fig. 9 of Ref. [34], where a near halving of laser absorption is predicted. What is more, the presence of non-Maxwellian EDFs also modifies heat transport and atomic kinetics processes, the full consequences of which are currently unknown [35–38]. It is important to note that the underlying motivation for many of the aforementioned studies is fusion research, and it is not known how the findings presented therein translate to lower-temperature, laser-driven extreme ultraviolet (EUV), or soft x-ray light sources. It is the goal of the present work to make strides in this direction.

In this paper, we investigate the Langdon effect in the realm of laser-driven EUV light source plasmas. We know of two studies, those of Sunahara *et al.* [8,39], that account for the aforementioned reduction in laser absorption in EUV source simulations. While Scott *et al.* [21] acknowledge that the Langdon effect will become important for state-of-the-art CO₂-driven (laser wavelength $\lambda_{\text{las}} = 10.6 \mu\text{m}$) plasmas [40], its impact on plasma properties and source performance was

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not quantified. Moreover, we are unaware of any studies that have investigated the consequences of non-Maxwellian EDFs on thermal transport and atomic kinetics in EUV source plasmas. Our goal is to bridge these gaps; we provide theoretical evidence that non-Maxwellian EDFs will emerge in EUV source plasmas and, furthermore, we calculate the corrections to laser absorption and thermal conduction as a result of these non-Maxwellian EDFs. In light of the complete absence of EDF measurements in EUV source plasmas, we restrict our analysis to the framework described by Langdon [27], and postpone an investigation of the effects of non-Maxwellian distributions on the Coulomb logarithm [34,41] (needed for deriving the laser absorptivity) to future work.

The Langdon effect can be summarized as a distortion of the EDF into a non-Maxwellian one due to the preferential heating of low-energy electrons [27]. The “strength” of this distortion is characterized by the Langdon parameter α , which represents the ratio of the IBA heating rate to the electron-electron collision rate:

$$\alpha = Z_{\text{eff}} \frac{v_{\text{osc}}^2}{v_{th}^2} = 3.735 \times 10^{-13} I_{\text{las}} \lambda_{\text{las}}^2 \frac{Z_{\text{eff}}}{T_e}. \quad (1)$$

Here, $Z_{\text{eff}} = \langle Z^2 \rangle / \langle Z \rangle$ is the effective ionic charge, v_{osc} is the oscillation velocity of an electron in the electric field of the laser, v_{th} is the electron thermal velocity, and T_e is the electron temperature in eV. If α is small [27], for instance, then electron-electron collisions are sufficiently frequent to “counteract” IBA heating and a Maxwellian distribution is maintained. Langdon obtained the following form for the reduction in laser absorptivity:

$$f_L = 1 - \frac{0.553}{1 + (0.27/\alpha)^{0.75}}. \quad (2)$$

You see here two important features. As $\alpha \rightarrow 0$ (electron-electron collisions dominate), $f_L \rightarrow 1$; near-100% absorptivity. The opposite trend, $\alpha \rightarrow \infty$ (IBA dominates), gives an upper limit to the reduction $f_L \approx 1 - 0.553 = 0.447$. With increasing α , the EDF deviates further and further from a Maxwellian. Matte *et al.* [28] found that these distributions are well described by super-Gaussian functions of the form $f_e(v) \propto \exp[-(v/v_m)^m]$, where m is the order of the super-Gaussian, $v_m^2 \equiv \frac{3\Gamma(3/m)}{\Gamma(5/m)} v_{th}^2$, and Γ is the gamma function. They presented a scaling relation between m and α of the form

$$m = 2 + \frac{3}{1 + 1.66/\alpha^{0.724}}. \quad (3)$$

In the limiting cases $\alpha \rightarrow 0$, $m \rightarrow 2$ (a Maxwellian distribution) and $\alpha \rightarrow \infty$, $m \rightarrow 5$. This scaling relation was found to be in good agreement with experimental measurements [29,31,32,42].

To determine f_L and m in EUV source conditions, we must first identify appropriate values of Z_{eff} , I_{las} , λ_{las} , and T_e . We know from atomic kinetics considerations (see Ref. [43] and the references therein) that the 13.5 nm-emitting tin charge states $Z \in [11^+, 15^+]$ are bred in plasmas with $T_e \in [20, 50]$ eV. We therefore “restrict” the ratio Z_{eff}/T_e , appearing in Eq. (1), to lie in the range $Z_{\text{eff}}/T_e \in [0.2, 0.6]$ eV⁻¹. Second, we know from the work

of Hemminga *et al.* [44] that the optimum laser parameters (that is, the laser parameters that generate highly efficient EUV-emitting plasmas) scale according to $I_{\text{las}} = 1.44 \times 10^{11} \lambda_{\text{las}}^{-1} \text{ W cm}^{-2}$ for $\lambda_{\text{las}} \in [1.064, 10.64] \mu\text{m}$, and thus $I_{\text{las}} \lambda_{\text{las}}^2 = 1.44 \times 10^{11} \lambda_{\text{las}} \text{ W cm}^{-2} \mu\text{m}^2$. As discussed in Hemminga *et al.* [44] and Gonzalez and Sheil [45], heating the plasma beyond the optimum laser intensity is a simple means of increasing EUV production. As a result, we fix $I_{\text{las}} \lambda_{\text{las}}^2$ to lie in the range $I_{\text{las}} \lambda_{\text{las}}^2 \in [10^{11}, 10^{13}] \text{ W cm}^{-2} \mu\text{m}^2$. While these ranges were derived with laser-driven tin plasmas in mind, the conclusions presented below are valid for a broader range of plasma-based light sources.

The results of our calculations for f_L and m are shown in Figs. 1(a) and 1(b), respectively. The colormaps extend from $f_L = 0.5$, $m = 4$ (yellow) to $f_L = 1$, $m = 2$ (blue) and the white lines represent contours of fixed f_L and m . First, we consider plasmas driven by short-wavelength Nd:YAG ($\lambda_{\text{las}} = 1.064 \mu\text{m}$) and Tm:YLF ($\lambda_{\text{las}} = 2 \mu\text{m}$) lasers [44,46–49]. For the former, we have $I_{\text{las}} \lambda_{\text{las}}^2 \sim 1.4 \times 10^{11} \text{ W cm}^{-2} \mu\text{m}^2$, giving $f_L \sim 0.95$ and $m \sim 2$; the reduction is rather minor, and the distributions remain close to Maxwellian. The Tm:YLF case is more interesting: $I_{\text{las}} \lambda_{\text{las}}^2 \sim 3 \times 10^{11} \text{ W cm}^{-2} \mu\text{m}^2$, giving $f_L \sim 0.85$ and $m \sim 2.3$. You see here a strong sensitivity of the reduction factor to even slight deviations of the EDF from a Maxwellian. This reduction is further enhanced for CO₂-driven plasmas, where typical irradiation conditions $I_{\text{las}} \lambda_{\text{las}}^2 \sim 10^{12} \text{ W cm}^{-2} \mu\text{m}^2$ gives $f_L \sim 0.8$ and $m \sim 2.4$. We should note that in our recent modeling of CO₂-driven plasmas [45], we did not account for this reduction in laser absorptivity, and this may contribute to differences between predicted and measured [40] conversion efficiencies. Regardless of the origin of this discrepancy, it is clear from the above that the Langdon effect should be incorporated into EUV source simulations.

It should also be remarked that the above analysis is based entirely on the frameworks of Langdon [27] and Matte *et al.* [28]. We do not consider, for example, the effect of atomic kinetics on the EDF as was investigated in the recent work of Le *et al.* [38]. Their inclusion of atomic kinetics processes in Vlasov-Boltzmann-Fokker-Planck (VBFP) simulations drove free electrons back toward a Maxwellian distribution, giving rise to a higher laser absorptivity ($\sim 10\%$) than predicted by Eq. (2). While we expect that atomic kinetics processes will have a similar effect in EUV source plasmas, this remains to be confirmed.

Thermal conduction is another critical plasma process modified by the Langdon effect. Transport theories, such as the classical Spitzer-Härm (SH) formalism [50], assume Maxwellian-distributed electrons, where the tail of the distribution ($v > 3v_{th}$) is primarily responsible for heat transport [51]. The Langdon effect is thought to depopulate this high-energy tail, giving rise to a lower heat flux than predicted by SH theory [51]. For example, a near-40% reduction of the SH thermal conductivity is expected for an $m \simeq 3$ super-Gaussian distribution [35,51]. When calculating the heat flux in such situations, an additional complication arises in that the order of the super-Gaussian, m , varies in space. This leads to two additional terms in the non-Maxwellian heat flux: one due to gradients in electron density, ∇n_e , and the other due to

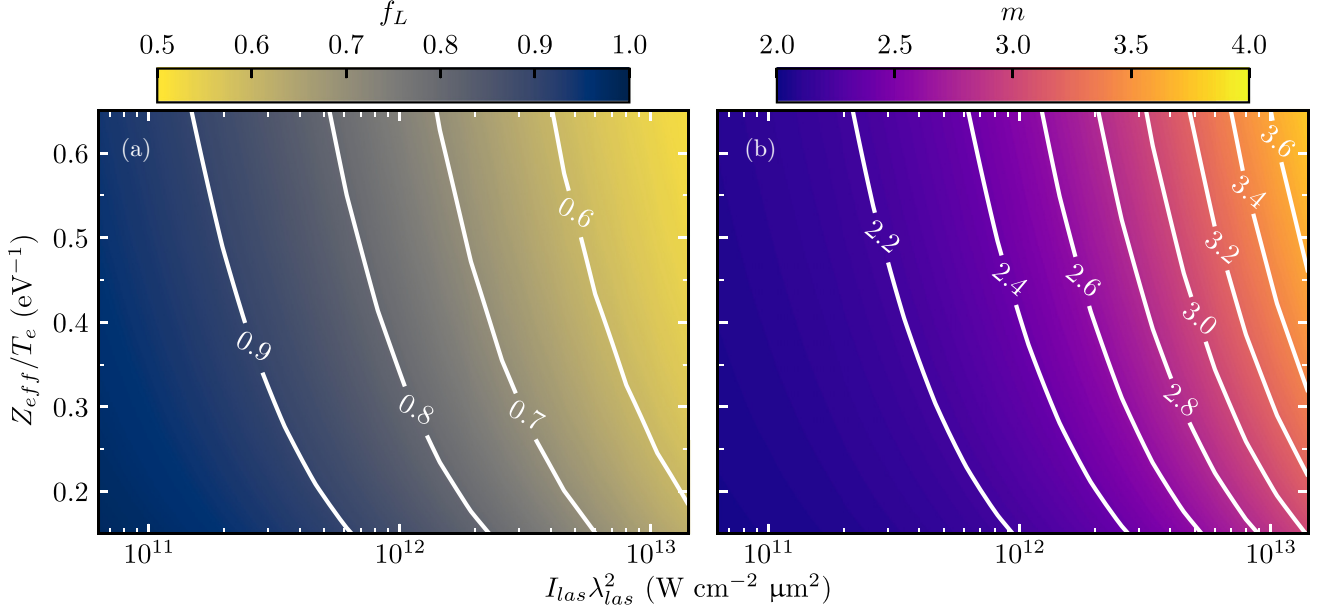


FIG. 1. (a) Langdon-induced reduction factor f_L and (b) super-Gaussian exponent m for the range of $I_{las}\lambda_{las}^2$ and Z_{eff}/T_e relevant for EUV source plasmas.

gradients in m , ∇m [35,52]:

$$Q_{non-Max} = -\left(C_T \kappa_{SH} \nabla T_e + C_n \kappa_{SH} T_e \frac{\nabla n_e}{n_e} + C_m \kappa_{SH} T_e \frac{\nabla m}{m}\right). \quad (4)$$

Here, κ_{SH} is the SH thermal conduction coefficient [50], and C_T , C_n , and C_m are correction factors due to non-Maxwellian EDFs. The correction due to the density gradient, C_n , is close to 0 for all m (see Fig. 1 of Ref. [52]), and thus we neglect

it in the following analysis. Expressions for C_T and C_m from Refs. [35,52] are

$$C_T = \frac{\Gamma\left(\frac{10}{m}\right)}{2} (7 - 5G)C, \quad (5)$$

$$C_m = \frac{\Gamma\left(\frac{10}{m}\right)}{2m} \left[35\psi\left(\frac{5}{m}\right) - 15\psi\left(\frac{3}{m}\right) - G\beta - 20\psi\left(\frac{10}{m}\right) \right] C, \quad (6)$$

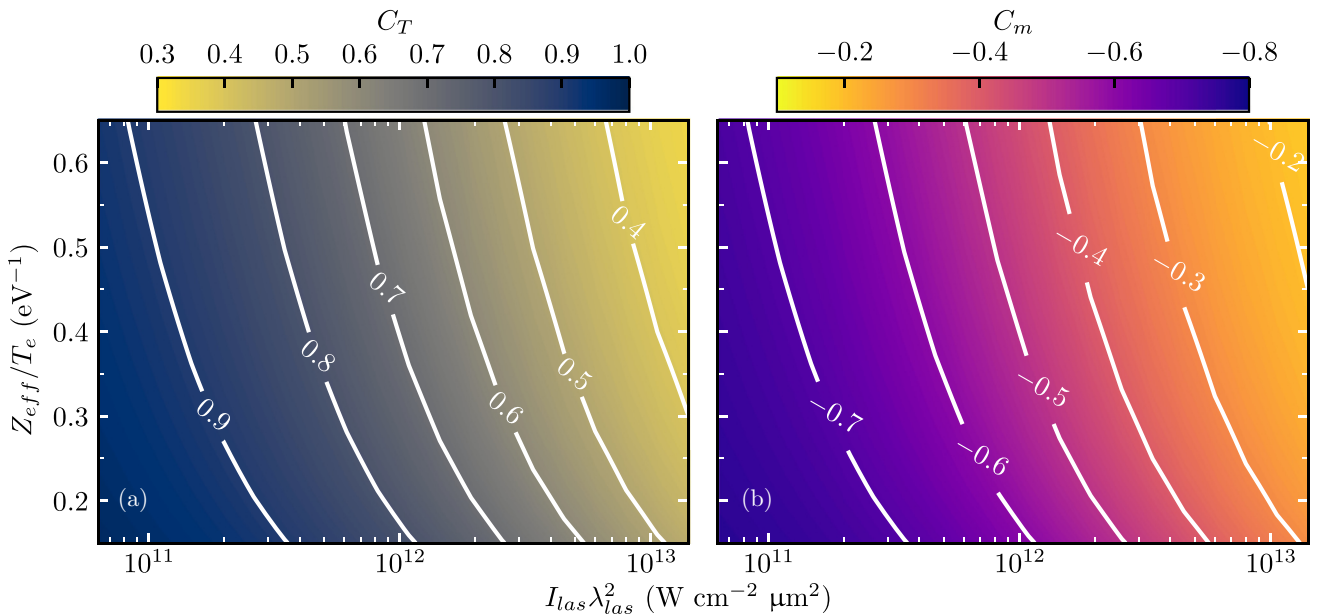


FIG. 2. Corrections to SH heat conductivity due to (a) temperature gradient (C_T) and (b) m gradient (C_m) in the relevant parameter range for LPP EUV sources.

where $C = \frac{\sqrt{\pi}}{48\Gamma(\frac{3}{m})} [\frac{3\Gamma(\frac{3}{m})}{2\Gamma(\frac{5}{m})}]^{\frac{1}{2}}$, $G = \frac{4[\Gamma(\frac{8}{m})]^2}{3\Gamma(\frac{6}{m})\Gamma(\frac{10}{m})}$, $\beta = 25\psi(\frac{5}{m}) - 9\psi(\frac{3}{m}) - 16\psi(\frac{8}{m})$, and $\psi(x) = \Gamma'(x)/\Gamma(x)$ is the polygamma function. All of these corrections depend on the plasma conditions implicitly through m . In Fig. 2, we present the corrections (a) C_T and (b) C_m , respectively, for EUV source conditions. Even for relatively low $I_{\text{las}}\lambda_{\text{las}}^2$ values, such as for Nd:YAG-driven plasmas, a near-10% reduction of the SH value is expected. These corrections are amplified for the CO₂-driven case: $C_T \sim 0.7$, a rather substantial correction. The correction C_m appearing in the ∇m term is everywhere negative and is similar in magnitude to C_T . Proper evaluation of these corrections in EUV source conditions requires proper modification of rad-hydro codes, such as that described in the ICF-relevant work of Huo *et al.* [35]. What is clear, however, is that the unique conditions associated with laser-driven EUV sources (that is, large values of Z_{eff}/T_e and laser wavelengths compared to ICF conditions) means that the Langdon effect can impact laser absorption and thermal conduction processes for laser intensities significantly lower than those typically used in ICF.

In conclusion, we have investigated the Langdon effect in the realm of EUV source plasmas. We have provided theoretical evidence that non-Maxwellian EDFs will emerge in these plasmas and modify both laser absorption and thermal conduction processes. It remains to be seen how these modifications alter our current understanding of laser-

driven EUV sources. Proper evaluation of the EDF in these conditions, such as through VBFP simulations, would be very insightful. Experimental confirmation of the presence of non-Maxwellian EDFs in EUV source plasmas is also highly desirable. For example, modifying the Thomson scattering setup of Tomita *et al.* [53,54] and Pan *et al.* [55] to something akin to Milder *et al.* [31] could provide new, fundamental insights on plasma-based light sources. Going further, laser-driven EUV sources may provide a more easily accessible platform (compared to ICF platforms) for testing and refining theoretical predictions of the Langdon effect and, moreover, its impact on laser absorption, thermal conduction, and atomic kinetics processes.

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