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Space charge corrected electron emission from an aluminum surface under non-equilibrium conditions

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A theoretical study has been conducted of ultrashort pulsed laser induced electron emission from an aluminum surface. Electron emission fluxes retrieved from the commonly employed Fowler-DuBridge theory were compared to fluxes based on a laser-induced non-equilibrium electron distribution. As a result, the two- and three-photon photoelectron emission parameters for the Fowler-DuBridge theory have been approximated. We observe that at regimes where photoemission is important, laser-induced electron emission evolves in a more smooth manner than predicted by the Fowler-DuBridge theory. The importance of the actual electron distribution decreases at higher laser fluences, whereas the contribution of thermionic emission increases. Furthermore, the influence of a space charge effect on electron emission was evaluated by a one dimensional particle-in-cell model. Depending on the fluences, the space charge reduces the electron emission by several orders of magnitude. The influence of the electron emission flux profiles on the effective electron emission was found to be negligible. However, a non-equilibrium electron velocity distribution increases the effective electron emission significantly. Our results show that it is essential to consider the non-equilibrium electron distribution as well as the space charge effect for the description of laser-induced photoemission. [http://dx.doi.org/10.1063/1.4729071]

I. INTRODUCTION

The interaction of short laser pulses with materials has been the subject of both fundamental and practical interests for years.[1–5] However, in spite of numerous efforts, the fundamental mechanisms of ultrashort laser ablation are still poorly understood.[6–10]

Laser induced electron emission has raised interest for several applications, such as photocathodes for free electron lasers and ultrashort x-ray pulses.[11–14] It has also been widely used to measure excited electron lifetimes and for the study of ultrafast electron dynamics in metallic surfaces, as well as surface states.[15–23]

Electron emission can affect the processes occurring during ultrashort ablation considerably.[24–26] A well known example is Coulomb explosion (CE), which would act as a potential material removal mechanism during the gentle phase of ablation, mainly for dielectrics.[27] This ablation mechanism consists of electrostatic disintegration of several atomic layers as a result of positive surface charging due to electron emission. The possibility of CE has been theoretically studied for metals, semiconductors, and dielectrics, assuming multi-photon photoemission (MPPE) and thermionic emission as primary mechanisms that induce charge separation in the target.[10] It was demonstrated that CE could act as a potential ablation mechanism in case of dielectrics, but not in case of metals or semiconductors. Aside from CE, it has been suggested that laser induced electron emission from a metallic surface could lead to early stage plasma formation.[28]

Ultrashort pulsed laser induced electron emission is commonly described by the Fowler-DuBridge (FD) theory.[10,27,29–34] It treats the total electron emission flux as the sum of the thermionic and the multiphoton photoelectron emission fluxes. The FD-theory assumes that the electrons in the metal are uniformly distributed in momentum space and obey Fermi-Dirac statistics. However, this assumption does not hold during and shortly after the laser pulse.[7,35–38] Since the majority of the electrons are emitted at this timescale, and the expressions in the FD-theory are derived as an integral over the thermalized electron distribution, a critical assessment of the Fowler-DuBridge theory seems necessary. As proposed in Ref. 39, both thermal and non-thermal effects of the electrons have to be taken into account. In this spirit, we apply the Boltzmann equation which provides insight into the temporal evolution of the non-equilibrium electron distribution. The calculated electron fluxes contain contributions from thermal as well as non-equilibrium electrons and will be compared to the fluxes calculated by the FD-theory.

Moreover, it has been suggested that electron emission can be significantly reduced by a space charge effect.[40–46] Since a large number of electrons are emitted in a short timescale, the negative charge outside the target generates an electric field that prevents electrons from being effectively emitted. Depending on the applied laser fluence, electron emission can be reduced by several orders of magnitude. The influence of the space charge effect should therefore also be

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considered in the comparison of both theories for ultrashort pulsed laser induced electron emission. This space charge effect could have a huge impact on various phenomena attributed to ultrashort pulsed laser induced electron emission, such as Coulomb explosion or early stage plasma formation.

A great deal of work has recently been devoted to the development of a more generally applicable theory for electron emission.[13,14,47] The focus lies here primarily on the combination of different theories in order to describe different regimes of laser induced electron emission. Furthermore, an external electric field is applied in order to increase the yield by counteracting the space charge effect and inducing field emission. These models are used to investigate applications such as photocathodes. Since our model does not consider the negative bias due to the external electric field, experimental validation is not an easy task. Instead, our work focuses directly on the non-equilibrium electron distribution and its effect on electron emission. We compare a commonly applied theoretical approximation for electron emission to a more advanced approach. In all cases, the space charge effect is considered. The latter was found to have a large influence on the net emitted electron flux.

II. THEORY

Calculations were performed for a perfectly homogeneous and isotropic aluminum sample. Laser pulses were chosen with a wavelength of 800 nm, for a pulse duration of 100 fs, and for fluences ranging from 3.32×10^{-3} J/m^2 to 1.33 J/m^2. These fluences should be regarded as absorbed fluences coupled directly to the target *after* reflection. The work function of Al is taken as 4.25 eV.[48]

A. Particle-in-cell (PIC) model

In order to describe the space charge effect on ultrashort laser induced electron emission for metals, a one dimensional PIC model was developed.[46,49] The PIC-model describes the emitted electrons as macroparticles with a certain velocity, position, and a certain weight to represent the number of electrons. These particles are then mapped onto a charge distribution via the cloud-in-cell method, which distributes the weight of each particle to its neighbouring gridpoints. The charge distribution is then used to calculate the local electric field by Gauss' law, assuming infinite parallel plates. This electric field can be interpolated to the particle positions in order to update their velocity and position by Newton's equation of motion. The calculation is depicted schematically in Fig. 1.

When particles reach the surface boundary they are pushed back into the solid, while at the remote boundary, they escape. Accordingly, the latter process can be identified as effective emission. The remote boundary is defined at a cut-off distance above the target. It is set far beyond the peak of the charge density distribution, here 500 nm above the target.

Employing the concept of infinite parallel plates, the remote boundary condition for the electric field can be estimated by the total emitted charge per area

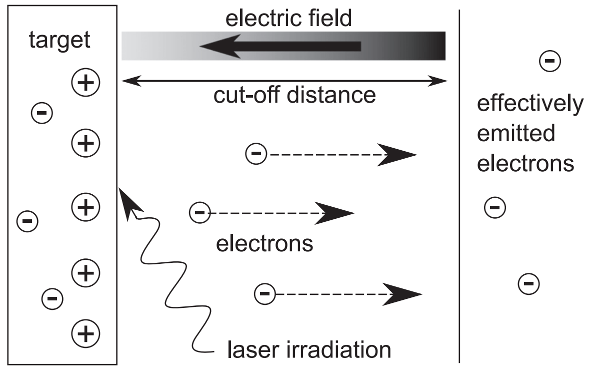

FIG. 1. A schematic representation of the model. The electrons in the target were modeled by the Boltzmann equation as well as by the FD-theory, the motion of emitted electrons in the computational area was modeled by the particle-in-cell code.

$$Q_{\rm em}(t) = \int_0^t J_{\rm em}(\tau)d\tau, \tag{1}$$

using the relation $E_{\rm rb}(t) = \frac{Q_{\rm em}(t)}{\epsilon_0}$.

B. Fowler-DuBridge theory

Particles are injected in the PIC-grid at the surface boundary. The number of particles that is injected in one timestep is commonly determined by the FD-theory.[46] This means that the total emission flux $J_{\rm tot}$ is a sum of the thermionic emission flux $J_{\rm therm}$ and the n-photon photoemission fluxes J_n

$$J_{\rm tot} = J_{\rm therm} + \sum_{n=1}^{\infty} J_n, \tag{2a}$$

$$= A_0 T_e^2 \exp\left(\frac{\phi}{k_B T_e}\right) + \sum_{n=1}^{\infty} a_n A_0 F(X_n) T_e^2 \left(\frac{e}{h\nu}\right)^n I^n, \tag{2b}$$

$$X_n = \frac{nh\nu - \phi}{k_B T_e}. \tag{2c}$$

The Fowler-function $F(X_n)$ can be evaluated as follows:

$$F(X_n) = \int_0^{\infty} \ln\left(1 + e^{-(y+X_n)}\right) dy, \tag{3a}$$

$$= \begin{cases} \sum_{n=1}^{\infty} (-1)^{n+1} \frac{e^{nX_n}}{n^2} & \text{if } X_n < 0 \\ \frac{\pi^2}{12} & \text{if } X_n = 0 \\ \frac{\pi^2}{6} + \frac{X_n^2}{2} - \sum_{n=1}^{\infty} (-1)^{n+1} \frac{e^{-nX_n}}{n^2} & \text{if } X_n > 0 \end{cases}. \tag{3b}$$

Other parameters are the Richardson-Dushman constant A_0, the electron temperature of the metal T_e, the work function ϕ, the Boltzmann constant k_B, the n-photon photoemission parameter a_n, the elementary charge e, the Planck constant h, the laser frequency ν, and the laser intensity I. The intensity is related to the fluence F_0 and the pulse duration τ via $I = F_0/\tau$, since we apply a square pulse.

C. Applying the Boltzmann equation for electron emission

In order to determine the electron flux, we use a kinetic approach to verify the FD-theory. In this work, the Boltzmann equation is applied to trace non-equilibrium electrons and phonons during and after the laser irradiation.[7] The scattering processes are determined by complete collision integrals. As in Rethfeld *et al.*,[7] we consider electron-electron and electron-phonon collisions as well as the absorption due to inverse Bremsstrahlung. As a result, the electronic distribution function can vary drastically from a thermalized Fermi-Dirac distribution. The electron emission fluxes are obtained upon integration of the actual non-equilibrium electron distribution

$$J_{\mathrm{tot}} = \frac{e}{4\pi^3 m\hbar^3}\int_0^\infty d\vec{p}\ \kappa(p_z) f[E(\vec{p})] p_z. \tag{4}$$

The parameters in this equation are the momentum normal to the surface p_z, the electron emission probability $\kappa(p_z)$, and the electron distribution $f[E(\vec{p})]$. The electron emission probability is assumed to be one for kinetic energies normal to the surface above the work function, and zero otherwise. We implemented the DOS for a free electron gas into the Boltzmann equation to determine the transient distribution function. Calculations are performed for a thin film of aluminum with a thickness much smaller than the mean free path of the hot electrons.[50] As a result, the target is homogeneously heated and spatial dimensions can be neglected.

Figure 2 shows a direct comparison of a non-equilibrium distribution during and after laser excitation and a thermalized Maxwellian tail of the Fermi distribution with the same internal energy and density. Since the work function is much higher than the thermal energy, defined by $k_B T_e$, the Maxwell-Boltzmann distribution provides a good approximation for the tail of the corresponding Fermi-Dirac distribution. Two figures are shown, for low and high fluences, respectively. For higher fluences, a faster thermalization is observed, see Fig. 2(b). This is due to a larger phase space the electrons can scatter in.[51] Obviously, non-equilibrium effects have a large impact on the electrons emitted shortly after laser excitation and therefore should be considered carefully.

D. Obtaining the Fowler-DuBridge parameters

The dependence of the photoelectric currents on the laser intensity in the FD-theory predicts that photoelectric emission would stop right after the laser pulse, see Eq. (2b), where the photoelectric currents are given by the second term. Instead, it is reasonable to assume that the emission would continue due to the thermalization of the non-equilibrium electrons. Another drawback of the FD theory is the *a priori* knowledge of the emission parameters a_n. According to our knowledge, these parameters have not yet been defined properly. Therefore, we apply the Boltzmann equation which is able to deal with thermal *and* non-equilibrium effects to calculate the emission fluxes. Subsequently, we extract an electron temperature from the non-equilibrium distribution by fitting a Fermi distribution obeying the same internal energy and density.[52] This is done to get input parameters for the FD-theory which is only defined in a thermal equilibrium assuming a Maxwell-Boltzmann distribution. The corresponding maximum temperatures, extracted from the energies of the non-equilibrium distributions, are shown in Fig. 3. As mentioned before, the work function is much higher than the thermal energy defined by $k_B T_e$, therefore, the Maxwell-Boltzmann distribution provides a good approximation for the Fermi-Dirac distribution.

The electron emission fluxes resulting from FD- and Boltzmann calculations, respectively, are shown for three different laser fluences in Fig. 5. They can be used to estimate the a_n-parameters. This is necessary for a consistent comparison of the Fowler-DuBridge theory with the fluxes calculated by the Boltzmann equation. Since the profiles of the emission fluxes are quite different for both theories, the emission parameters a_n should not be estimated from the transient fluxes. Instead, it is more realistic to base the a_n-parameters on the total emitted charge. Hence the a_n-parameters were estimated by scaling the total emitted charge per area derived from the equilibrium FD-fluxes to the ones retrieved from the non-equilibrium electron distribution. At low levels of excitation, the three photon emission process

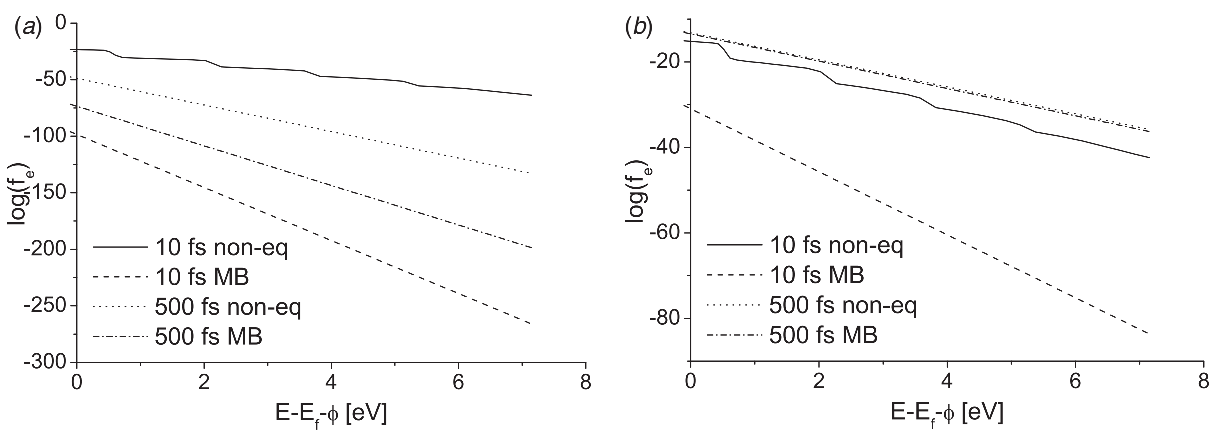

FIG. 2. The tails of the electron distributions for energies above the work function, during and after laser irradiation with a fluence of (a) $F_0 = 0.05\,\mathrm{J/m^2}$ and (b) $F_0 = 0.85\,\mathrm{J/m^2}$. The labels non-eq and MB denote non-equilibrium distribution calculated by the Boltzmann equation, and a Maxwell tail of the Fermi distribution based on the corresponding internal energy and density, respectively.

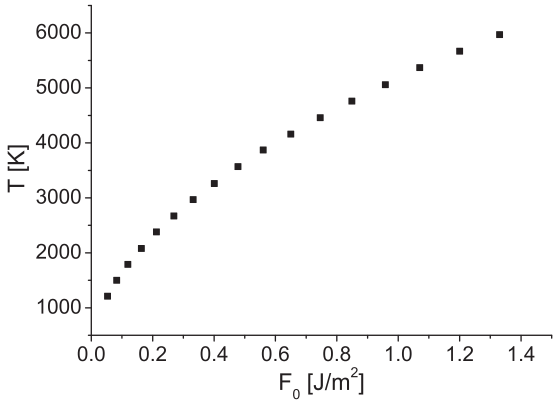

FIG. 3. The peak electron temperature corresponding to the energy distribution for simulations with different laser fluences.

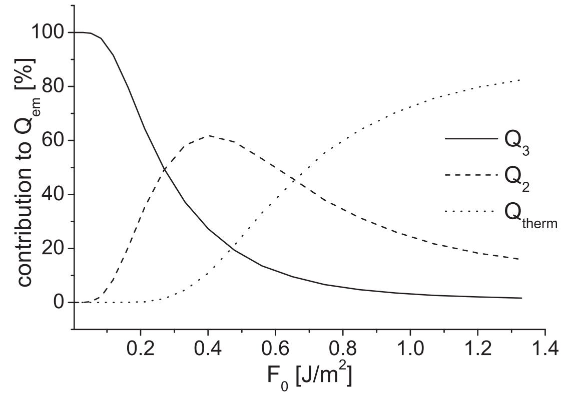

FIG. 4. The relative contributions of the 3PPE, 2PPE, and thermionic emission process, denoted by Q_3, Q_2, and Q_{therm}, respectively, to the total emitted charge in percentages, as a function of the laser fluence.

(3PPE), dominates. However, at higher levels of excitation the two photon emission process (2PPE) will become more important, because at higher temperature, a higher fraction of electrons would need to absorb less energy to cross the energy barrier of the work function.[53] Accordingly, calculations were performed for a large range of fluences. At low fluences, 3PPE is found to be dominant. Therefore, we estimated a_3 by scaling the total emitted charge per area as calculated by the FD theory at low fluences, to the one calculated by the Boltzmann equation. The next parameter a_2, can then be derived for moderate fluences by deducting the resulting total emitted charge per area by 3PPE calculated by the FD theory, and scaling the result to the total emitted charge per area. For high fluences, thermionic emission dominates and the resulting emission flux can be predicted by the Richardson-Dushman equation. For a_3, a value of $1.24 \times 10^{-44}\,\mathrm{m}^6/\mathrm{A}^3$ was extracted from calculations for a laser fluence between $3.32 \times 10^{-3}\,\mathrm{J/m}^2$ and $0.163\,\mathrm{J/m}^2$, for a_2 the value was fitted to $3.28 \times 10^{-29}\mathrm{m}^4/\mathrm{A}^2$, comparing calculations for laser fluences from $0.269\,\mathrm{J/m}^2$ to $0.478\,\mathrm{J/m}^2$. These values are valid for the assumed wavelength of the laser and the density of states for a free electron gas. Upon the implementation of just thermionic emission, 2PPE and 3PPE, the total charge corresponds well with the one calculated from the non-equilibrium electron distribution. Hence, the 1PPE process cannot be separated from the other emission processes for our conditions, since thermionic emission takes over at fluences above the 2PPE regime.

III. RESULTS AND DISCUSSION

A. Fowler-DuBridge theory

The implementation of the a_n-parameters derived above, results in a good agreement between the total emitted charge derived from the FD-theory and the Boltzmann equation ($<10\%$) for laser fluences below $1.33\,\mathrm{J/m}^2$. The relative contributions to the total emitted charge of the different emission processes conform to the FD-theory. In Fig. 4, the relative contributions to the total emitted charge Q_x/Q_{tot} are illustrated, respectively (see Eq. (2)).

As previously stated by Anisimov,[53] the 3PPE process dominates at low laser intensities, since three photons are necessary to cross the energy barrier of the work function. From a laser fluence of approximately $0.212\,\mathrm{J/m}^2$, a higher fraction of electrons will need less energy to be emitted due to the higher level of excitation, so the 2PPE process becomes important. At even higher levels of excitation (from a laser fluence of approximately $0.75\,\mathrm{J/m}^2$), solely the kinetic energy of a larger fraction of electrons will be enough to be emitted. The latter can be considered as thermionic emission. The transition from MPPE to thermionic emission as obtained from our model is in the same fluence range as observed in experiments.[54,55]

The electron emission fluxes calculated by the FD-theory and based on the Boltzmann equation considering non-equilibrium effects are depicted in Fig. 5.

As mentioned in Sec. II D, for relatively low fluences, the resulting electron emission flux is quite different for the FD-theory as compared to the one based on the non-thermal electron distribution. The main flaw of the FD-theory stems from its strict dependence on the laser intensity. Consequently, the FD-theory predicts relatively high electron emission at the start of the laser pulse, though it is clear that the electrons have to undergo a finite excitation process in order to be emitted. Similarly, the FD-theory predicts that emission stops abruptly after the laser pulse (see the second term in Eq. (2b)), while the calculation with the Boltzmann equation reveals that non-thermal emission continues for some time. It can also be seen that at higher laser fluences, thermionic emission gains importance, and that thermionic emission is relatively well described by the commonly accepted Richardson-Dushman equation. This reflects that at higher laser fluences, the electron distribution thermalizes more quickly than at low intensities,[7] since the Richardson-Dushman equation assumes that the electrons follow a thermalized distribution (see also Fig. 2). This result is confirmed by a thermalization time $\tau \propto 1/T_e^2$, as predicted by the Fermi-liquid theory.[51]

B. Space charge effect

The space charge effect on ultrashort pulsed laser induced electron emission is modeled by a one-dimensional PIC-code. At the target, surface electrons are emitted and

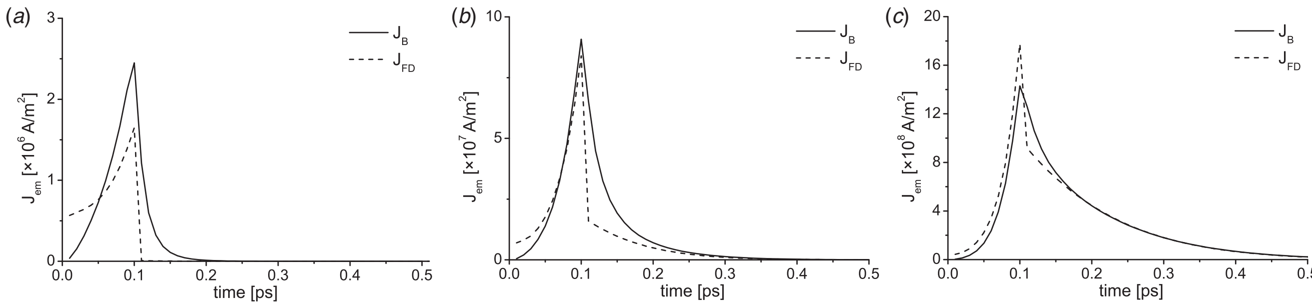

FIG. 5. The electron emission pulse shape resulting from the Boltzmann equation (solid) and calculated by the FD-theory (dashed) for laser fluences of 0.212, 0.478, and 0.849 J/m^2.

enter the computational domain. Here, the amount of emitted electrons is defined by the electron emission flux, while the velocity distribution is derived from the energy distribution. Simulations were performed for three types of input: The first consists of a flux calculated by the FD-theory, using Maxwellian velocity distributions, the second employs a flux derived from the solution of the Boltzmann equation but assuming a Maxwellian velocity distribution, whereas the third applies both the flux and velocity distribution based on the non-equilibrium electron distribution, resulting from the Boltzmann approach.

The results for the effective emission yield Q_{em}, i.e., the effectively emitted electrons per area are shown in Fig. 6. One would conclude from the analysis of the Maxwellian cases that the FD theory provides an adequate description of the space charge corrected laser induced electron emission. However, the velocity distribution *does* make a significant difference. The non-equilibrium velocity distribution results in a higher fraction of electrons emitted with a higher initial velocity (see Fig. 2). This results in an increased effective emission yield. For higher fluences, the difference between the three input-sets is less pronounced. Thermionic emission is dominant at these high levels of excitation and the influence of the photo-emitted currents becomes negligible. Moreover, the electrons will thermalize quicker at high fluences (see Fig. 2), which reduces the non-equilibrium effects in the considered time scale. Apparently, here the Richardson-Dushmann equation provides an adequate description of the initial electron emission since it is based on a thermalized electron distribution.

The relative yield, i.e., the ratio of the number of effectively emitted electrons to the total number of emitted electrons is shown along with the maximum values for the electric field above the target in Fig. 7.

The space charge corrected emission yield is similar employing equilibrium or non-equilibrium based initial fluxes. But the higher initial velocity in the non-equilibrium case causes more electrons to be effectively emitted. It is also clear that at higher fluences a reduced yield is observed due to the increased electric field above the surface.

It is interesting to note that even at low fluences, the space charge effect reduces electron emission significantly. Assuming a non-equilibrium electron distribution, the electron emission is already reduced by 25% for a laser fluence of 0.21 J/m^2. In absolute numbers, electron emission is quite limited at these fluences. Assuming a spot size diameter of 10 μm, less than 1000 electrons would be effectively emitted. This indicates that for the major part of the fluence regime, the space charge effect must be considered when quantifying electron emission.

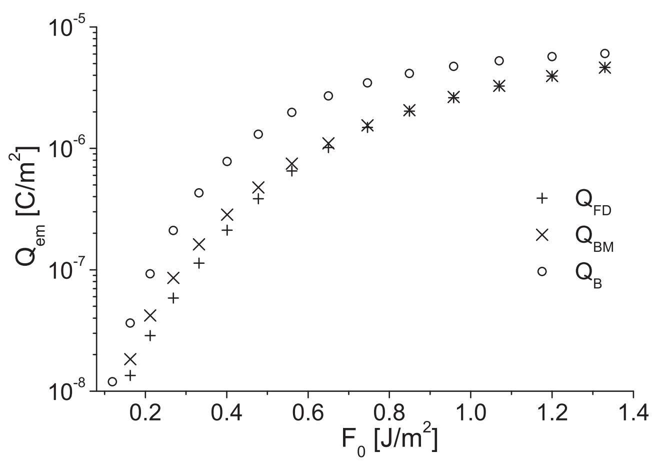

FIG. 6. The number of effectively emitted electrons as a function of the laser fluence, calculated by PIC-simulations assuming an electron flux calculated by the FD-theory, and a Maxwellian velocity distribution (FD), an electron emission flux taking into account the actual electron distribution and a Maxwellian velocity distribution (BM) and an electron emission flux as well as a velocity distribution based on the actual electron distribution (B).

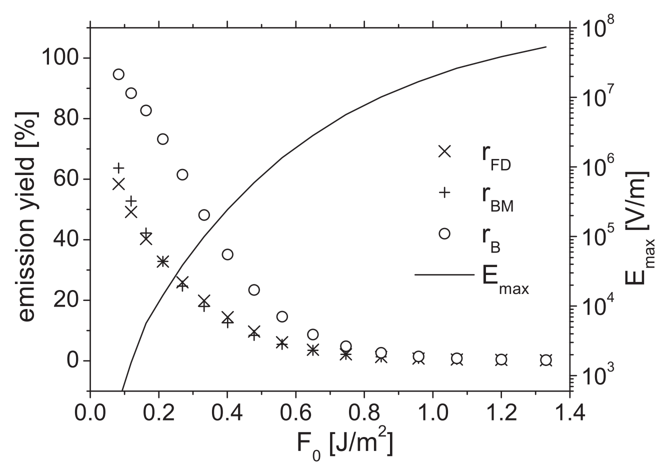

FIG. 7. The yield of effectively emitted electrons relative to the initially emitted charge Q_{init} as a function of the laser fluence, including the space charge effect for different laser fluences. The labels are as in Fig. 6. The solid curve shows the maximum value of the electric field above the target for each fluence.

IV. CONCLUSION

Ultrashort pulsed laser induced electron emission was studied, considering the non-thermal electron distribution during and after the laser pulse. We presented a method to determine the FD-parameters a_n, and calculated them for our specific case. It was found that the Fowler-DuBridge theory is not valid at low fluences where multiphoton photoelectron emission is dominant. Besides, the influence of the transient electron distribution must be considered. This is not surprising, since the FD theory is based on the Maxwell-Boltzmann distribution while the initial equilibrium distribution is significantly modified when a metal is irradiated by an ultrashort laser pulse. For higher fluences, the Richardson-Dushman equation provides an adequate description for laser induced thermionic emission. This is a consequence of the faster thermalization of the electrons for higher excitations.

Furthermore, the space charge effect was found to reduce the effective electron emission considerably. It starts already at low fluences and increases as higher fluences applied. Therefore, it is essential to consider a space charge in models describing electron emission.

While the fluxes calculated by the FD-theory are quite different compared to those calculated from the non-equilibrium distribution, the difference in the flux shape has a minor influence on the space charge corrected electron emission. However, the velocity distribution *does* make a significant difference. In the Boltzmann approach considering non-equilibrium excitation, more electrons are effectively emitted due to the higher initial velocity as compared to the Maxwellian assumption. The effective emission flux is influenced more by the velocity distribution than by the transient electron flux shape. Despite the fact that the electron fluxes can be approximated by the Fowler-DuBridge theory, a kinetic model is indispensable to determine the laser affected electron velocity distribution, strongly affecting the total emitted charge.

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