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Scattering and the relationship between quantum efficiency and emittance

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Simple models of the quantum efficiency (QE) and emittance ($\varepsilon_{n,rms}$) of metals are based on the neglect of scattered electrons to the emission current. The leading order terms in the Fowler-Dubridge equation for QE and the Dowell-Schmerge equation for emittance entail $QE \propto \varepsilon_n^4$. Here, a method to account for next-order contributions and the impact of scattered electrons to the relation is given.
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In vacuum electronics and advanced light sources, the quest for high brightness electron sources engenders that the principal performance metrics are current density and emittance.[1,2] For photocathodes, current density is tantamount to quantum efficiency (QE), which is the ratio of number of emitted to photoexcited electrons, and its maximization is important; conversely, emittance is a measure of the tendency of a beam to diverge as it propagates, and its minimization is likewise important.[3–5]

As shown by Fowler[6] and Dubridge (FD),[7] QE is proportional to $U[\beta(\hbar\omega - \phi)]$ for metals, where $U(x) \approx x^2/2 + \pi^2/6$ is the Fowler function,[8] $\beta = 1/k_BT$, $\hbar\omega$ is the photon energy, and $\phi = \Phi - \sqrt{4QF}$ is the height of the emission barrier above the Fermi level μ for the image charge potential, Φ is the work function, $Q = 0.36$ eV nm, and F is the product of the electric field and elementary charge in units of [eV/nm]. The term $\sqrt{4QF}$ is also known as the Schottky factor. At room temperature, the $x^2/2$ factor dominates, suggesting $QE_{FD} \propto (\hbar\omega - \phi)^2$ (as also shown by the three step model of Spicer[9,10] and the moments approach[11–13]). Using similar approximations to FD, Dowell and Schmerge (DS)[14] showed that the transverse emittance is given by $\varepsilon_{n,rms} \propto (\hbar\omega - \phi)^{1/2}$ to leading order (a relation that holds well even when temperature and field effects are included[13]). In tandem, these relations suggest that $QE \propto \varepsilon_{n,rms}^4$ (the "DS relation" below). Simultaneous measurements of QE and $\varepsilon_{n,rms}$ for simple metals exhibit the relationship for small $\hbar\omega - \phi$ (see, for example, the data for Antimony (Sb) by Vecchione in Ref. 15, but which also show departures at higher energies from the simple relation). As argued by Dowell and Schmerge, the connection between QE and $\varepsilon_{n,rms}$ is of special interest given the dependence of the former on the longitudinal momentum $\hbar k_z$ and the latter on the transverse momentum $\hbar k_\perp$ of the photoexcited electron. Because QE and $\varepsilon_{n,rms}$ rise and fall together, how they do so is important as an improvement in one is to the detriment of the other.

The FD and DS equations neglect the contribution of scattered electrons, as is common (see, though, Ref. 9 which considers singly scattered electrons in a complex analysis). In contrast, semiconductors like GaAs, with its negative electron affinity (NEA) surface, show temporal response behavior (Figure 9 of Ref. 16) that shows the contribution of scattered electrons to the yield.[17] A cesiated metal surface can exhibit a very low Φ, thereby significantly increasing the probability that scattering events need not be fatal to photoemission from metals.[11,18] Therefore, we shall (i) reconsider the relation between QE and $\varepsilon_{n,rms}$ using a moments approach, (ii) find the contribution of scattering, and (iii) compare the analytic findings to a numerical evaluation for conditions associated with cesiated surfaces.

In the moments formulation, the mean value $\langle A \rangle$ of $A(k_j) = k_j^n$ for $n =$ an integer power and $j =$ a Cartesian index (i.e., x, y, z, or $\perp$) is obtained by integration over the emission distribution. Letting $k = |\vec{k}|, E = (\hbar k)^2/2m$, then $A_{QE} = k_z = k\cos\theta$, whereas $A_\varepsilon = k_\perp^2 \equiv (k^2 + k_\omega^2)\sin^2\theta$, where $k_\omega^2 = 2m\omega/\hbar$. When the transmission probability is approximated by a step function in energy, electrons will not be emitted unless $(E + \mu)\cos^2\theta \geq \mu + \phi$, implying that the maximum angle $\theta_{max} \equiv \varphi$ of the "escape cone" is $\sin^2\varphi = (\hbar\omega - \phi)/(\hbar\omega + \mu)$ where μ is the electrochemical potential (also referred to as the Fermi level - the notation and parameters follow Ref. 18). The "fatal approximation" assumes that scattered electrons are eliminated and introduces $f_\lambda(E) = \cos\theta/(\cos\theta + p(E + \hbar\omega)) \rightarrow s/(s + p)$, where $\cos\theta \equiv s$, to account for their removal. For photoexcited electrons in metals, $p(E) = \delta/l(E) \approx p_o(u - 1)^\eta/\sqrt{u}$ for $u = E/\mu$ and $p_o = 6.22$ and $\eta = 1.9$ for Cu (copper), where the laser penetration depth $\delta \approx 12$ nm.[13] For $1/k_BT$ large, the Fermi-Dirac distribution is well-approximated by a Heaviside step-function.

In keeping with a Monte Carlo analysis, the electron population is characterized by an expanding shell of unscattered electrons plus a sphere of scattered electrons.[18] As a dome-like portion of the shell passes the surface, the average longitudinal velocity of the *emitted* electrons decreases and the average transverse velocity increases—while less important for fast response emitters like metals, the observation is consequential for semiconductors where δ is larger and the emission time greater, and will matter when the pulse length of a photoexcited bunch is on the order of several picoseconds. After a sufficient time has elapsed, the mathematical formulation of the Shell model becomes equivalent to the DS approach.

The leading order QE behavior is now found as follows. Observing that $QE \propto \langle k_z \rangle$, performing the azimuthal integration, and letting $\cos\theta = s$ gives

$$\langle k_z\rangle = \frac{\int_{k_m}^{k_F} k^2 dk \int_{\cos\varphi(k)}^{1} ds[ks^2/(s+p)]}{\int_{k_o}^{k_F} k^2 dk \int_{-1}^{1} ds[1]}, \tag{1}$$

where $\hbar^2 k_m^2/2m = \mu - (\hbar\omega - \phi)$ and $\hbar^2 k_o^2/2m = \mu - \hbar\omega$. The s integrations are analytic. The odd usage of $[\cdots]$ in the integrands is to motivate the notation

$$\langle k_z\rangle \equiv \frac{\hat{O}[ks^2/(s+p)]}{\hat{O}_o[1]}, \tag{2}$$

where the "o" subscript in the denominator means setting the lower bound of the s-integration to –1 instead of $\cos\varphi$ in the denominator of Eq. (1). The $\hat{O}$ notation is defined by comparing Eqs. (2) to (1) and used exclusively below. When $\hbar\omega - \phi$ is small $p(E)$ weakly varies. Letting $p(E+\hbar\omega) \rightarrow p(\mu+\hbar\omega)$, the integrals become analytic. The dominant contribution to the angular integration in the numerator is proportional to $\sin^2\varphi$. To leading order, then, $QE \propto (\hbar\omega - \phi)^2$.

The leading order $\varepsilon_{n,rms}$ behavior is similarly found as follows. For uniform emission from a circular area (laser radius spot size R_c), $\varepsilon_{n,rms} = (\hbar/mc)\sqrt{\langle x^2\rangle\langle k_x^2\rangle}$. When R_c greatly exceeds the distance between scattering events, $\langle x^2\rangle = R_c^2/2$. Alternately, the common *mean transverse energy* (*MTE*) notation, as in $\varepsilon_{n,rms} = R_c\sqrt{MTE/mc^2}$, is used to avoid reference to R_c, and *MTE* is easily related to $\langle k_x^2\rangle$. Therefore, speaking of $\sqrt{MTE}$ is equivalent to speaking of $\varepsilon_{n,rms}$, particularly when ratios are taken so that common factors are eliminated. As the denominator is over *photoemitted* electrons, f_λ appears in the denominator, and so

$$\langle k_x^2\rangle \equiv \frac{\hat{O}[(k^2+k_\omega^2)(1-s^2)s/(s+p)]}{\hat{O}[s/(s+p)]} \tag{3}$$

and so the denominator uses $\hat{O}$, not $\hat{O}_o$. The dominant contribution to the $s-$integration is proportional to $\sin^4\varphi$ in the numerator, but $\sin^2\varphi$ in the denominator. To leading order, then $\langle k_x^2\rangle \propto (\hbar\omega - \phi)$. Taken with the QE result, it implies $QE \propto \varepsilon_n^4$.

The "dominant contribution" parlance can be understood as follows. From the identity

$$1 = \frac{\cos\theta}{1+p} + \frac{p}{1+p} + \frac{\sin^2(\theta/2)}{1+p}, \tag{4}$$

the terms are like the fraction of unscattered (first term) plus scattered (second term) electrons and a negligible (third) term: the FD and DS equations are then seen to assume that the first term of Eq. (4) dominates. Exercising greater precision, the quantities f_λ and $(1-f_\lambda)$ are in fact the proportion of scattered and unscattered electrons, respectively. For the $(1-f_\lambda)$ electrons, f'_λ of them will likewise suffer no additional scatterings before they are emitted, whereas $(1-f'_\lambda)$ will suffer an additional scattering, where $f'_\lambda = f_\lambda(E')$ and E' is the post-scattering electron energy. For iterated scatterings,

$$1 = f_0 + \bar{f}_0\{f_1 + \bar{f}_1[f_2 + \bar{f}_2(\ldots(f_n + \bar{f}_n))]\}, \tag{5}$$

where $\bar{f}_j = 1 - f_j$ and n indicates the number of scatterings endured until emission is precluded. For metals e–e scattering dominates, followed by acoustic photon: although the latter scatters isotropically and results in a minor energy change, the former aggressively removes energy from the photoexcited electron and forbids directions that would allow final states below the Fermi level.[18] For a cesiated surface ($\Phi \approx 1.6$ eV) and incident blue or UV light ($\hbar\omega > 3.5$ eV) the first scattering event is mostly isotropic, and that approximation will be made. Although sufficiently energetic electrons can share their energy with a Fermi-level electron such that both are capable of emission, it is assumed that on average only one of them travels towards the surface. Thus, the proportion of scattered electrons capable of emission after j events satisfies $\bar{f}_1\,\bar{f}_2\ldots\bar{f}_n \gtrsim \prod_{j=1}^{n} p_j/(1+p_j) \equiv P_n$, an important approximation as the factor can be removed from the integrals and a lower limit established if p_j is approximately constant and evaluated at the reduced energy of the scattered electron. If the scattered electrons equally share the energy of the primary colliding electrons, then $E_{j+1} = E_j/2 = \mu + \hbar\omega/2^j$ by $E_0 = \mu + \hbar\omega$ and mathematical induction, an austere approximation to enable an analytic final form. For metals, an electron that is emitted after two scattering events is rare, so $j \leq 1$ (in contrast, for semiconductors, optical phonons dominate such that $E_j \approx \hbar\omega - E_g - j\Delta$, where Δ is the phonon energy and E_g is the band gap: the analysis consequently becomes more involved), and so

$$\hat{O}[A] \rightarrow \hat{O}[A]_0 + P_1\hat{O}[A]_1 \tag{6}$$

in Eqs. (2) and (3), where $[A]_j$ is taken to mean $p = p(E_j)$ if and when it occurs in A. A similar equation holds for $\hat{O}_o$. Close inspection reveals that this is tantamount to $QE \rightarrow QE_0 + P_1 QE_1$ with $\hbar\omega \rightarrow \hbar\omega/2^j$ in QE_j. The denominator in Eq. (3) complicates $\varepsilon_{n,rms}$: if the numerator of Eq. (3) is $\hat{O}[A]$ and the denominator $\hat{O}[B]$, then

$$\frac{\hat{O}[A]}{\hat{O}[B]} \rightarrow \frac{\hat{O}[A]_0 + P_1\hat{O}[A]_1}{\hat{O}[B]_0 + P_1\hat{O}[B]_1} \tag{7}$$

from which the impact on $\varepsilon_{n,rms}$ is found. Clearly, scattering will cause a departure from the DS relation, for which the final task is to estimate its scattering-infused form. That shall be done by numerically performing the integrals entailed in $\hat{O}[A]$.

Two cases are considered: first, for conditions characteristic of bare metals, in which DS is the presumptive leading order term and second, for cesiated metals where differences are expected. For bare metals (Copper-like: $\mu = 7$ eV, $\Phi = 4.5$ eV, $F = 10$ eV/μm), then $\hat{O}[A]/\hat{O}[B] = \hat{O}[A]_0/\hat{O}[B]_0$ for both QE and $\varepsilon_{n,rms}$, as expected, but a better representation of the relationship between them is given by $QE(\lambda)/QE(\lambda_o) = x^4/(\bar{a} + ax^b)$, where $\bar{a} = 1-a$ and $x = \varepsilon(\lambda)/\varepsilon(\lambda_o)$, $a = 0.0379$, $b = 2.385$, and $\lambda_o = 275$ nm (approximately ϕ expressed as a wavelength). The new relationship is suggested by demanding that for $x \lesssim 1$, DS is recovered, but for $x > 1$ there is a transition to a different power law behavior. DS is good within 95% for $x \leq 1$.

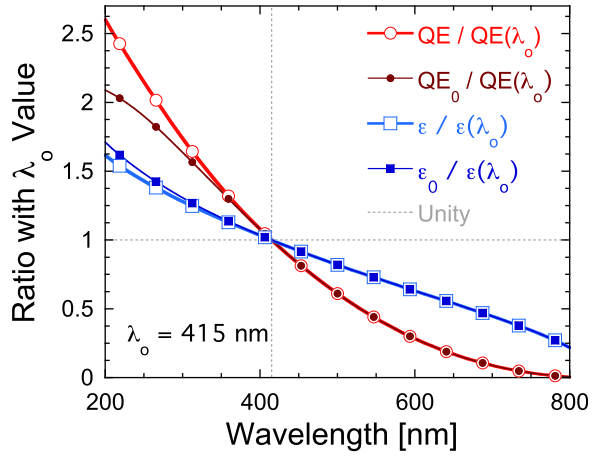

FIG. 1. Ratios of QE (red dots) and ε (blue squares) with reference terms evaluated at $\lambda_0 \approx 2\pi\hbar c/\phi$ (ratios eliminate multiplicative factors).

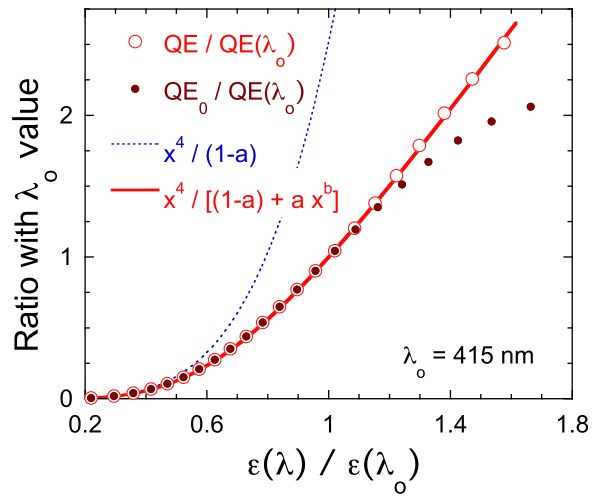

FIG. 2. Ratios of QE (open dot) and QE_0 (solid dot) with $QE(\lambda_o)$ as functions of $\varepsilon/\varepsilon(\lambda_o)$. Blue dashed line $\propto$ DS relation. Red solid line evaluated using $a = 0.6029$ and $b = 2.67$.

For cesiated metals using copper-like parameters ($\mu = 7\,\text{eV}$, $\Phi = 1.6\,\text{eV}, F = 10\ \text{eV}/\mu\text{m}$), then the relationships are, as expected, more complex. First, a direct comparison of QE and $\varepsilon_{n,rms}$, along with QE_0 and ε_0, are shown in Fig. 1 for $\lambda_o = 415$ nm, followed by an assessment of the DS and fitted form in Fig. 2. The fit to $x^4/(\bar{a} + ax^b)$ gives $a = 0.6029$ and $b = 2.67$. The DS relation is now good within 95% for $x < 0.4$, and the effects of scattering are more pronounced when $x > 1$.

In conclusion, the DS relation between QE and $\varepsilon_{n,rms}$ has been reassessed for cesiated surfaces and the contribution to scattering (both in $p(E)$ and in contributions to emission) in their relation quantified. It has been shown that before scattering contributes, the DS power law relation is modified to a form given by $x^4/(\bar{a} + ax^p)$ (that is, the denominator is different than unity in the reassessed DS relation) apart from other factors that affect experimental relations.[15] As a result, the trade-off between QE and $\varepsilon_{n,rms}$ as metrics of beam quality for metals is altered when scattering effects are explicitly included. Finally, it is conjectured that the analogous relations for semiconductors will be *more* consequential given that the phonon-mediated energy losses from scattering are weaker.

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