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Increase of intrinsic emittance induced by multiphoton photoemission from copper cathodes illuminated by femtosecond laser pulses

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Electron sources driven by femtosecond laser have important applications in many aspects, and the research about the intrinsic emittance is becoming more and more crucial. The intrinsic emittance of polycrystalline copper cathode, which was illuminated by femtosecond pulses (FWHM of the pulse duration was about 100 fs) with photon energies above and below the work function, was measured with an extremely low bunch charge (single-electron pulses) based on free expansion method. A minimum emittance was obtained at the photon energy very close to the effective work function of the cathode. When the photon energy decreased below the effective work function, emittance increased rather than decreased or flattened out to a constant. By investigating the dependence of photocurrent density on the incident laser intensity, we found the emission excited by pulsed photons with sub-work-function energies contained two-photon photoemission. In addition, the portion of two-photon photoemission current increased with the reduction of photon energy. We attributed the increase of emittance to the effect of two-photon photoemission. This work shows that conventional method of reducing the photon energy of excited light source to approach the room temperature limit of the intrinsic emittance may be infeasible for femtosecond laser. There would be an optimized photon energy value near the work function to obtain the lowest emittance for pulsed laser pumped photocathode. © 2018 Author(s).

Metallic photocathodes are used for high brightness electron beams with applications such as the free electron laser (FEL)¹ and ultrafast electron diffraction (UED)²,³ due to their prompt temporal response and robustness to poor vacuum conditions. The ultimate performances, such as the lower limit of the FEL output wavelength⁴ and the upper limit of the unit cell dimension of the UED sample,⁵ and the cost of these systems are affected deeply by the transverse emittance of electron beams. The intrinsic emittance is also becoming the lowest possible limit of a beam’s emittance with the introduction of emittance compensation process.⁶

The normalized transverse emittance of an electron beam can be expressed as

\[ \varepsilon_{n,x} = \frac{\sigma_x \sqrt{\langle p_x^2 \rangle}}{m_e c}, \]

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The normalized transverse emittance of an electron beam can be expressed as

\[ \varepsilon_{n,x} = \frac{\sigma_x \sqrt{\langle p_x^2 \rangle}}{m_e c}, \]
where $\sigma_x$ is the RMS (root mean squared) beam size, which can be replaced by the laser spot size on the cathode for the intrinsic emittance, $p_x$ is the transverse momentum of the electrons in $x$ direction, $m_e$ and $c$ are the electron rest mass and the speed of light, respectively. Accordingly, there are two methods to reduce the intrinsic emittance: focusing the laser spot smaller, and reducing the RMS transverse momentum of the emitted electrons. The minimum of the spot size is limited by the quality of the light source and laser beam control. Moreover, for a certain charge intensity, too small spot will induce large current density, which brings space charge effect. In addition, when more than a small number of electrons is needed, the low quantum efficiency of metals requires high laser intensities. Then lowering the laser spot size to reduce emittance risks pitting the photocathode surface. With an isotropic assumption at 0 K, the RMS transverse momentum is determined by the incident photon energy $\hbar \omega$ and the effective work function of the photocathode $\phi_{\text{eff}}$,

$$\sqrt{\langle p_x^2 \rangle} = \sqrt{\frac{m_e (\hbar \omega - \phi_{\text{eff}})}{3}}.$$  

Therefore, one can reduce the transverse momentum by tuning the laser photon energy to match the effective work function.\textsuperscript{8–10} J. Feng \textit{et al.}\textsuperscript{11} extended the theory and showed that the smallest achievable intrinsic emittance was limited by the lattice temperature of cathode as the incident photon energy approached the work function. Those prior measurement were taken with picosecond pulses or continuous light.

This letter reports the measurement of intrinsic emittance of a copper cathode illuminated by femtosecond laser pulses at photon energies above and below the work function of the cathode. The measured intrinsic emittance consists with theories and other’s experiments at higher photon energy above the work function. However, when the photon energy decreased below the effective work function, emittance increased rather than decreased or flattened out to a constant. We found that the photoemission excited by photons with sub-work-function energy contained two-photon photoemission that increased the emittance. This finding is useful for the choice of photon energy used to excite a practical photocathode and beneficial for the understanding of the emission mechanism of metal cathode driven by femtosecond pulses.

To measure the transverse intrinsic emittance of a photocathode, we built an apparatus (See details of the apparatus in the supplementary material) based on free expansion method.\textsuperscript{12,13} Its physical construction is shown in FIG. 1. The ultrashort laser pulses used in our experiment were generated from a wavelength tunable Ti: sapphire infrared laser, from which the pulse duration was less than 100 fs (FWHM), and 3rd harmonic generator system. The repetition rate of the infrared laser was 80 MHz. Passing through a vacuum viewport (not shown in the illustration) and the coated (for electrical conduction) glass, the pulsed laser was focused to about 100 $\mu$m FWHM spot onto the cathode at 35° incidence angle. The laser power was adjusted, with UV fused silica reflective neutral density filters, low enough to ensure that the number of electrons per pulse was equal to (corresponding to a photocurrent of 12.8 pA) or less than one averagely. For different wavelengths from 25 fJ to 12 pJ

FIG. 1. Mechanical model of measuring apparatus.
of optical pulse energy was used to complete the emittance measurement. Thus, the space charge effect was excluded. Photoelectrons emitted from the sample were accelerated in the uniform electric field between the cathode and anode in parabolic trajectories. More than half of the electrons passed through the grid and drifted along straight lines in the field free range before coming across the scintillator screen where they were imaged and recorded by a monochrome CCD camera. The grey value of a point on the image depends linearly on the number of electrons arrived at the corresponding point of the scintillator screen. The electron’s transverse momentum \( p_x \) at the sample surface is proportional to its displacement \( x \) from the center of the spot on the screen:

\[
p_x = \frac{x}{2g + d} \sqrt{2m_e eV},
\]

where \( g = 5 \text{ mm} \) is the gap between the sample and the grid, \( d = 250 \text{ mm} \) is the distance from the grid to the scintillator screen, \( V = 5 \text{ kV} \) is the absolute value of the potential applied on the cathode, and \( e \) is the charge of an electron. Obviously, the RMS transverse momentum \( \sqrt{\langle p_x^2 \rangle} \) is proportional to the RMS displacement \( \sqrt{\langle x^2 \rangle} \) with the same coefficient \( \sqrt{2m_e eV/(2g + d)} \).

To obtain the transverse intrinsic emittance from an electron beam image, e.g., the inset in FIG. 2, the following processes should be performed: (1) finding the center of beam spot; (2) calculating the average grey values of all the pixels with equal distance to the center of the spot to reduce the random noise, e.g., the green thick line in FIG. 2; (3) fitting these data with a double Gaussian function

\[
f(x) = \frac{A}{B} \exp \left( -\frac{x^2}{2B^2} \right) + \frac{C}{D} \exp \left( -\frac{x^2}{2D^2} \right) + E,
\]

where \( E \) represents the uniform background of the images; (4) formulating the RMS displacement \( \sqrt{\langle x^2 \rangle} \) with fitted parameters \( A, B, C, \) and \( D \), and \( \sqrt{(AB^2 + CD^2)/(A + C)} \). Finally, the transverse intrinsic emittance \( \varepsilon_{n,x} \) is expressed as

\[
\varepsilon_{n,x} = \frac{\sigma_x}{(2g + d)c} \sqrt{\frac{2eV}{m_e}} \sqrt{\frac{AB^2 + CD^2}{A + C}}.
\]

This form of the fitting function Eq. (4) is obtained by attempting, with which the adjusted coefficient of determination of the fitted curves were nearly one (>0.9998) for all the images involved in this paper and the relationship between emittance and fitting parameters is not too complex. For the polycrystalline cathode, we assumed an isotropic emission. Moreover, the influence of the laser spot shape was negligible owing to its quite small size (about 100 \( \mu m \) FWHM) compared to the beam radius at the scintillator screen, millimeters. The rotational symmetry of the spot, insert in FIG. 2,
FIG. 3. Measured transverse emittance (red squares) for polycrystalline copper cathode and the laser intensity (blue circles) used during the emittance measurement at different photon energies. Laser intensity is calculated as the pulse energy divided by the product of laser spot area and the pulse duration (FWHM). The laser spot area is indicated by the region where the brightness is greater than \(1/e^2\) times the maximum value. Different laser intensities were selected to hold the electron numbers of single pulse roughly constant (between 0.6 and 1). The effective work function is 4.09 eV.

on the screen also exhibited that the distribution in any direction was the same and could be used to calculate the transverse intrinsic emittance.

The transverse intrinsic emittance of a fine polished (mirror-like) polycrystalline oxygen-free copper cathode was measured at different photon energies (FIG. 3) by the method above. For the photon energies from 4.2 eV to 4.9 eV typically used in copper photocathode electron gun, the values of transverse intrinsic emittance increased from 0.4 \(\mu m/mm\) to 0.6 \(\mu m/mm\). The measured values were in good agreement with other experiments and theoretical models. The emittance decreased with the reduction of photon energy for energies higher than 4.10 eV, which was also consistent with previous results. However, the emittance increased abnormally rather than flattens out to a thermal limited constant when photon energy was further reduced. A minimum emittance value was found to be of 0.39 \(\mu m/mm\) at the photon energy 4.10 eV that was nearly the measured effective work function of the cathode (4.09 eV) according to Fowler’s theory (See obtainment of the effective work function in the supplementary material). The accuracy of measured values would be influenced by the finite laser spot size and the defocus effect due to of the anode grid holes, which make the measured emittance larger. By adjusting the laser spot size and selecting the appropriate grid, the effects of these two factors on the measured values are less than 0.5% and 2% (See two main error sources in the supplementary material).

The increase of transverse intrinsic emittance should result from several reasons, such as surface roughness, laser heating, and multiphoton photoemission. The laser spot positions on the cathode did not obviously change for different photon energies in our measurement. Moreover, the laser intensities (<1 MW/cm\(^2\)) used here were far below the melting threshold of copper (larger than tens of GW/cm\(^2\) for sub-ps pulses). Therefore, the surface roughness remained nearly the same for measurement at different photon energies. Further, according to numerical calculation from the two-temperature model, the electron temperature increase of the cathode due to the laser pulse with a maximal intensity was only about 1 K (see the numerical calculation of the electron temperature in the supplementary material), which did not account for emittance increase at lower photon energy range.

To confirm if multiphoton photoemission processes occurred, we observed the dependence of photocurrent density on incident laser intensity at specific photon energy shown in FIG. 4(a). The red line is a linear fit of the red squares for a higher photon energy 4.26 eV while the blue curve is a parabolic fit of blue dots for a lower photon energy 3.86 eV. We also plotted the photocurrent density as a function of the laser intensity in logarithmic coordinates and made linear fits like that in FIG. 4(b). The slopes (see the explanation about the fitted slopes in the supplementary material) of fitted lines for lower photon energies shown in FIG. 5 were all obviously larger than one, which indicated that the photoemissions were not pure single-photon process at the photon energies close to or lower than the effective work function of the cathode. The increase of fitted slope with the
FIG. 4. (a) Current density, charge of a single pulse divided by the product of laser spot area and the pulse duration (FWHM), for two different photon energies as a function of laser intensity. (b) Current density vs laser intensity in the log-log scale. The slopes of the fitted lines (solid lines) show notable difference. According to Eq. (6), for a pure n-photon photoemission, the dependence of photocurrent density on the laser intensity is a line with an integral slope in the log-log plot. Therefore, a non-integral slope indicates mixed multiphoton processes.

reduction of photon energy suggested a decrease of single-photon photoemission current. Therefore, we considered that the process contains multiphoton (mostly two-photon) photoemission.

The introduction of multiphoton photoemission should raise the measured emittance values. According to the generalized Fowler-Dubridge theory, the total photocurrent density $J$ is the sum of n-photon photoemission current density $J_n$,

$$J = \sum_{n=0}^{\infty} J_n = \sum_{n=0}^{\infty} \sigma_n I^n,$$

(6)

where $I$ is the incident laser intensity, and $\sigma_n$ is the generalized n-photon ionization cross section as a function of the photon energy, the electronic temperature, and the properties of the photocathode. For $n = 0$, $J_0 = \sigma_0$ corresponds to the Richardson’s equation for the thermionic emission. Obviously, the measured mean squared transverse momentum is the weighted mean of n-photon photoemission mean squared transverse momentum $\langle p_x^2 \rangle$,

$$\langle p_x^2 \rangle = \sum_{n=0}^{\infty} \frac{J_n}{J} \langle p_{x,n}^2 \rangle.$$

(7)

Based on the theory in Ref. 7, the RMS transverse momentum $\sqrt{\langle p_x^2 \rangle}$ of a n-photon photoemission ($n \geq 2$) would be expressed like Eq. (2) with $h\omega$ replaced by $nh\omega$, which would result in a remarkable increase of $\langle p_x^2 \rangle$ and hence the transverse intrinsic emittance. However, for pure n-photon process,

FIG. 5. Linear fitting slope of current density vs laser intensity in the log-log scale (red squares) and two-photon photoemission current ratio (blue asterisks) as a function of photon energy.
the emittance should decrease with the reduction of photon energy. There should be a trade-off in determining the variation tendency of transverse intrinsic emittance between the reduction of photon energy and the change of ratio of multiphoton photoemission current to the total photocurrent, which is the key point in study the abnormal increase of transverse intrinsic emittance.

By fitting the current density data of lower photon energy with a polynomial to obtain the coefficient $\sigma_n$, the ratio of the n-photon photoemission current to the total photocurrent $J_n/J$ can be calculated by

$$J_n/J = \frac{\sigma_n I^n}{\sum_{m=0}^{\infty} \sigma_m I^m}. \quad (8)$$

$\sigma_0$ was set to be zero for the reason that the electron temperature was close to room temperature during the measurement. In addition, when $n \geq 3$, $J_n$ becomes very small relative to the cases $n = 1$ or 2. Therefore, we used a parabolic curve with intercept equals 0,

$$J = \sigma_1 I + \sigma_2 I^2, \quad (9)$$

to fit these data, like the blue curve in FIG. 4(a), and got the values of $\sigma_1$ and $\sigma_2$ at different photon energies. Then the ratio of the two-photon photoemission current to the total photocurrent $J_2/J$ was calculated with the laser intensity used for the emittance measurement (blue asterisks in FIG. 5). It was found that the ratio of the two-photon photoemission current increased from nearly zero to about 34% while the photon energy reduced from 4.18eV to 3.86eV. Furthermore, the emittance component induced by two-photon photoemission should be much larger than that of single-photon photoemission electrons, which flattens out to a constant. Accordingly, the measured mean emittance of single and two-photon photoemission would increase with the reduction of the photon energy. Therefore, we primarily attributed the abnormal increase of emittance to the proportion increase of the two-photon photoemission in total emission processes.

According to Eq. (6), Eq. (7), and Eq. (1), the emittance should depend on laser intensity. For several specific photon energies, we measured respectively the emittance with different laser intensities, FIG. 6. The emittance has no connection with the laser intensity when the excited photon energy was higher than the work function. However, when the cathode was excited by lower photon energy laser, the emittance increased with the laser intensity for the introduction of multiphoton process and the ratio of multiphoton emission current increased with laser intensity. In our measurement, different laser intensities (blue circles in FIG. 3) were selected for different photon energies to ensure that the electron numbers of single pulse were between 0.6 and 1. In practical applications, the laser intensity should be determined by the required electron beam intensity and the gain of the cathode. A lower photon energy corresponding to a lower gain usually requires higher laser intensity, and then the ratio of two-photon photoemission will increase, resulting in an increase of emittance.

FIG. 6. Measured transverse emittance for several specific photon energies at different laser intensities. The average electron numbers per pulse are between 0.2 and 0.8.
Therefore, for femtosecond pulses, it may be infeasible to reduce the intrinsic emittance to room temperature limit\(^{11}\) by decreasing the photon energy lower than the work function of photocathode. There would be an optimized photon energy value near the work function to obtain the lowest emittance for pulsed laser pumped photocathode, which may extend to applications of other kinds of photocathodes.

In conclusion, we investigated the intrinsic emittance of copper photocathode illuminated by femtosecond pulses as a function of photon energy based on free expansion method. We obtained a result consistent with others’ at higher photon energy, but the intrinsic emittance increased when the photon energy decreased near and lower than the work function of the cathode. The result could be interpreted by the introduction of two-photon photoemission.

**SUPPLEMENTARY MATERIAL**

See supplementary material for (1) details of the apparatus, (2) the obtainment of the effective work function, (3) two main error sources, (4) the numerical calculation of the electron temperature, and (5) the explanation about the fitted slopes.

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