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MEASUREMENTS OF BARIUM PHOTOCATHODE QUANTUM YIELDS AT FOUR EXCIMER LASER WAVELENGTHS

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ABSTRACT

The electron quantum yields from barium cathodes excited by excimer laser radiation at 193, 248, 308 and 351 nm have been determined. Experiments with different cathode surface preparation techniques reveal that deposition of barium film a few micron thick on a clean copper surface under moderate vacuum conditions achieves relatively high quantum efficiencies. Quantum yields measured from surfaces prepared in this manner are 2.3×10^{-3} at 193 nm, 7.6×10^{-4} at 248 nm, 6.1×10^{-4} at 308 nm, and 4.0×10^{-4} at 351 nm. Other preparation techniques, such as laser cleaning of a solid barium surface, produced quantum yields that were at least an order of magnitude lower than these values.

Extensive research has been conducted recently on the use of photocathodes to produce short pulses of electrons.[1-4] These short pulse length sources have important applications in free-electron lasers, high-power microwave generation, and ion sources. A quantum yield (defined as the number of electrons emitted per incident photon) of 6.2 x 10^{-4} has recently been reported for magnesium irradiated by a Nd:YAG laser at 266 nm [5]. Since barium has a similar electronic structure and has a work function, $\phi_{Ba} \approx 2.3\,eV$ [6], which is even lower than that of magnesium, $\phi_{Mg} = 3.66\ eV$ [5], it may have an even greater quantum efficiency. This makes barium an extremely attractive candidate for use as a photocathode material.

A major challenge in using barium photocathodes is the need to keep the surface free from contaminants. Barium is an extremely reactive element which readily adsorbs and reacts with even trace amounts of water vapor and oxygen. This paper describes three possible methods for creating an efficient cathode surface for photoemission applications: precleaning under an inert atmosphere, cleaning at low pressures with high laser fluences, and getter wire deposition of barium onto a copper substrate under high vacuum conditions. Of these, deposition of a fresh barium surface was found to produce the highest quantum yield. Photoemission from barium prepared by this technique was studied at four excimer laser wavelengths.

The experimental setup for this experiment is illustrated in Figure 1. A 22 cm diameter stainless steel vacuum chamber pumped by a cryosorption pump houses the photocathode. The typical base pressure in the chamber is 2 x 10^{-7} torr. The cathode is mounted on the end of a copper stem extending into the center of the chamber at a 45° angle to the incident laser beam. This cathode holder is connected to electrical ground through a picoammeter. The collection anode, a graphite disk which can be biased up to 3 kV, is located in the chamber approximately 32 mm from the cathode surface. Excimer laser radiation at 193, 248, 308, or 351 nm with a pulse width of approximately 20 ns and a repetition rate of 3-10 Hz is used to excite the photocathode, stimulating electron emission. The laser beam is directed through an iris and onto a series of glass plates. Reflections from these plates are used to attenuate the laser beam. The beam enters the chamber through a quartz window. With this configuration, the irradiated

cathode area can be varied between 5 mm^2 and 30 mm^2 and the incident energy can be reduced to $\leq$ 1 μJ/pulse. Laser pulse energies were measured using a pyroelectric detector. Quantum yield measurements were corrected for the transmission of the window.

Photo-induced current is detected by two methods. A picoammeter attached to the cathode grounding line measures the average photocurrent. Additionally, a current transformer with a time response of approximately 10 ns is connected to the anode bias line, producing a direct measurement of the pulse width of the photoelectrons. The latter arrangement is essential in order to verify that only photoemission, rather than thermionic emission, of electrons is occurring. Thermionic emission would last much longer than the laser pulse.

Two types of cathodes were tested: a vacuum spray-deposited thin barium film and a solid barium cathode. Within the vacuum chamber is a moveable holder on which fifteen 1 cm-long segments of barium getter wire are mounted. The getters contain an alloy of barium, aluminum and nickel which releases barium vapor when heated to 1000° C. Barium is deposited on the cathode surface (a copper substrate) by resistively heating the getter wires. This requires approximately 11 amperes per wire segment. Pressure in the chamber before and during deposition is $< 5 \times 10^{-7}$ torr. After two minutes of heating, the current is shut off and the getter assembly is moved away from the coated cathode. According to the manufacturer's data, 1 cm of getter wire should yield approximately 1.5 mg of barium. Based on this, the 15 wire segments used should generate approximately 25 mg, which would produce a layer of barium 40 μm thick on the copper cathode. Assuming that 20% of the theoretically emitted barium actually strikes and adheres to the cathode surface, one deposition with this assembly would produce a layer 8 μm thick which would be equivalent to about 20,000 monolayers of barium atoms. Rough measurements of the mass of barium deposited on the cathode were made with an analytical balance. Measured values were on the order of 2 mg of barium, yielding 3 μm thick layers. A few experiments were performed which had two getter assemblies in the vacuum vessel. This allowed the preparation of films twice as thick as the normal layers without opening the chamber.

Solid barium surfaces were prepared by melting barium granules on a copper disk in an inert gas atmosphere. After cooling, the cathodes are stored under vacuum-degassed liquid paraffin. (This paraffin is a liquid at room temperature.) Just prior to use, the cathode surfaces are cleaned with sandpaper under more paraffin, rinsed with high purity degassed hexane, and transferred to the chamber. These steps all occur in an inert gas atmosphere. For some experiments the solid cathodes were subjected to an additional preparation step, consisting of irradiating the emitting area with high power UV laser pulses while under high vacuum.

Quantum yields were measured for barium excited at 193, 248, 308 and 351 nm by an excimer laser. Irradiation of evaporation-deposited barium surfaces at these wavelengths yielded quantum efficiencies of 2.3×10^{-3}, 7.6×10^{-4}, 6.1×10^{-4}, and 4.0×10^{-4} respectively. Figure 2 is a plot of these results. Also shown are the quantum efficiencies obtained for the uncoated copper substrate. As can be seen, the barium photocathode has a much higher quantum efficiency than copper at all wavelengths. At 308 nm, the barium is more than 4 orders of magnitude better than the copper. These high quantum efficiencies of the deposited barium layers are also fairly stable over time. At a chamber base pressure of 2×10^{-7} torr, the quantum efficiency drops by only 25% during the first half of an hour after deposition. Further decreases occured more slowly, with many cathodes retaining more than half their efficiency 50 hours after preparation.

The effectiveness of the three cathode surface preparation methods were compared using 308 nm light. Experiments with the solid barium photocathode cleaned only by surface abrasion produced quantum efficiencies of less than 1×10^{-6}.. The cathode surface was grey rather than the silver-white color characteristic of pure barium, indicating that the surface was exposed to oxygen or water vapor during the pump-down period. Though Anderson and Hunt [7] reported that the work function of barium does not change upon exposure to small amounts of oxygen, that work only tested this effect under very controlled conditions. The barium surface they tested was coated by less than 100 monolayers of oxygen atoms. It also did not study the effect of water, another possible contaminant in the present experiment. Barium will react with water

to form barium hydroxide. In this experiment, the solid cathodes are handled in either deoxygenated liquid paraffin, hexane, or in an inert argon atmosphere. Residual levels of O_2 and H_2O were not measured, though, and it is quite likely that trace amounts of these contaminants remained in these environments, causing photocathode degradation.

Though the untreated solid barium cathode was an inefficient photoemitter, the quantum yield was substantially increased by laser cleaning the surface with excimer laser radiation. Young et al [8] reported that quantum yields from solid barium cathodes could be increased by a factor of ten after irradiation by 308 nm laser light at a laser fluence of 30 mJ/cm^2. They proposed that this enhancement occurs either by thermal desorption of contaminants or plasma formation which cleans the surface. In the present work, laser fluences up to 700 mJ/cm^2 were used to clean the barium. A quantum yield of 1 x 10^{-4} for 308 nm light was obtained after 5 minutes of laser cleaning at 1.25 Hz with this intensity. Increasing the cleaning time did not improve the quantum efficiency. Higher laser fluences also did not improve the quantum efficiency. At the higher fluences, black burn marks formed on the cathode surface. Damaged cathode surfaces had quantum efficiencies of less than 5 x 10^{-5}.

To alleviate the difficulties associated with preparing a clean, unoxidized barium surface for photoemission measurements, cathode surfaces were prepared by depositing barium from heated getter wires onto a clean copper substrate. This technique has several advantages. The getter wires are relatively unaffected by exposure to air and thus require no special handling. The cathode surface is prepared at pressures below 5 x 10^{-7} torr. As a result, contamination by atmospheric gases is minimized. Additionally, the sprayed barium which is not deposited on the cathode target coats the walls of the vacuum chamber where it can act as a getter to remove trace amounts of oxygen, water or other reactive gases which might contaminate the cathode surface. The reported peak quantum yields for barium at each of the tested wavelengths were measured using cathode surfaces prepared by this method. No visible degradation was observed after the laser irradiation. Using two getter assemblies in the vacuum chamber allowed for the preparation

of films which were twice as thick as normally used. Quantum yields from these "double-thick" layers are the same as those from the usual thickness layers.

Extensive research has been conducted on the photoemissive properties of thin films of barium deposited on tantalum, tungsten, and quartz[6,9,10]. In Gaudart's work [6], the work function showed a sharp decrease for films on the order of 3-4 nm thick, but then increased to 2.3 eV for thicker films. This result was also reported by Shul'man *et al* [9] who found a slight minimum in work function corresponding to a monolayer thickness of barium on their tungsten surface. The barium layers deposited in the current experiment are much thicker than this, (on the order of 6-10 μm), so they should behave much as the bulk metal would. Because of this, the deposited layers should be more rugged than thin films. This fact is verified by the ability of the cathode to withstand relatively high laser intensities (700 mJ/cm$^{2)}$ without any decrease in quantum efficiency. In addition, deposition of more than one layer of barium on the cathode did not affect the quantum yield.

The factor of eight increase in quantum yield from 351 to 193 nm indicates that even greater efficiencies might be possible with higher energy photons. To determine what benefit could be gained by using shorter wavelength exciting light, the experimental data was fitted to the empirical equation proposed by Fowler [11].

$$\log\left(I/T^2\right) = B + \log(f[(h\upsilon - \chi)/kT])$$

where

$$f[x] = e^x - \frac{e^{2x}}{2^2} + \frac{e^{3x}}{3^2} - \ldots \qquad x \le 0$$

$$\frac{\pi^2}{6} + \frac{x^2}{2} - \left(e^{-x} - \frac{e^{-2x}}{2^2} + \frac{e^{-3x}}{3^2} - \ldots\right) \quad x \ge 0$$

I is the photocurrent, T is the temperature, χ is the work function, and B is an arbitrary constant. The results of this fit using T=293.15 K and χ=2.5 eV are shown in Fig 3. This expression was derived for the wavelength region around the photoemission threshold and is not strictly valid for wavelengths far from this threshold, which is 2.5 eV for barium. However, as can be seen, the data is at least qualitativly fit by the prediction. If the extrapolation to the larger photon energies

is correct, a factor of three increase in the quantum yield may be achievable if 150 nm light is employed rather than 193 nm light, the shortest wavelength used in this study. These short wavelengths suffer from increased attenuation by the atmosphere and many window materials, however; it may be more desirable to increase the number of photons, i.e. increase the laser power, rather than the energy per photon in order to produce more electrons from the photocathode.

In summary, the electron quantum yields of barium excited by excimer laser radiation were measured over the range 351 to 193 nm. Peak efficiencies were obtained using cathodes prepared by depositing a 6-10 μm layer of barium onto a copper substrate from commercially available getter wires. Photocathodes prepared by this method had quantum yields as high as 2.3×10^{-3}. The quantum efficiency of these surfaces is relatively stable, with some cathodes exhibiting quantum yield half lifes in excess of 50 hrs at an ambient pressure of $1\text{-}2 \times 10^{-7}$ torr. This confirmation of high efficiency and stability makes barium an attractive candidate for short electron pulse operation applications.

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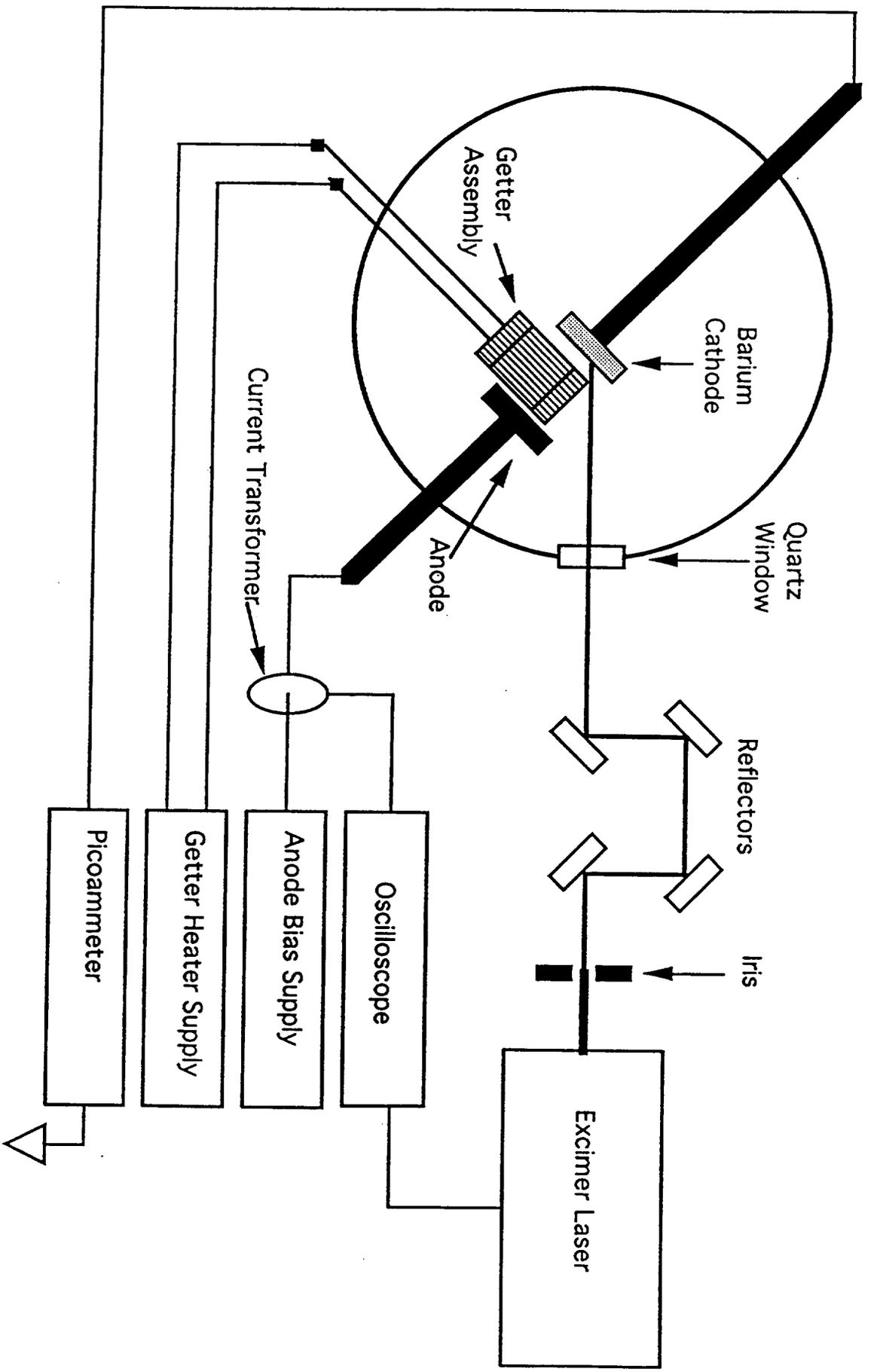
FIGURE CAPTIONS

FIGURE 1. Diagram of experimental configuration. Getter assembly is mounted so that it can be moved out of the laser path during photocurrent measurements.

FIGURE 2. Plot of quantum efficiencies for copper and barium measured at 193, 248, 308 and 351 nm.

FIGURE 3. Plot of experimenally determined barium quantum yields and Fowler's theoretical curve for a material with barium's work function.


Getter Assembly
Barium Cathode
Quartz Window
Anode
Current Transformer
Reflectors
Iris
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Oscilloscope
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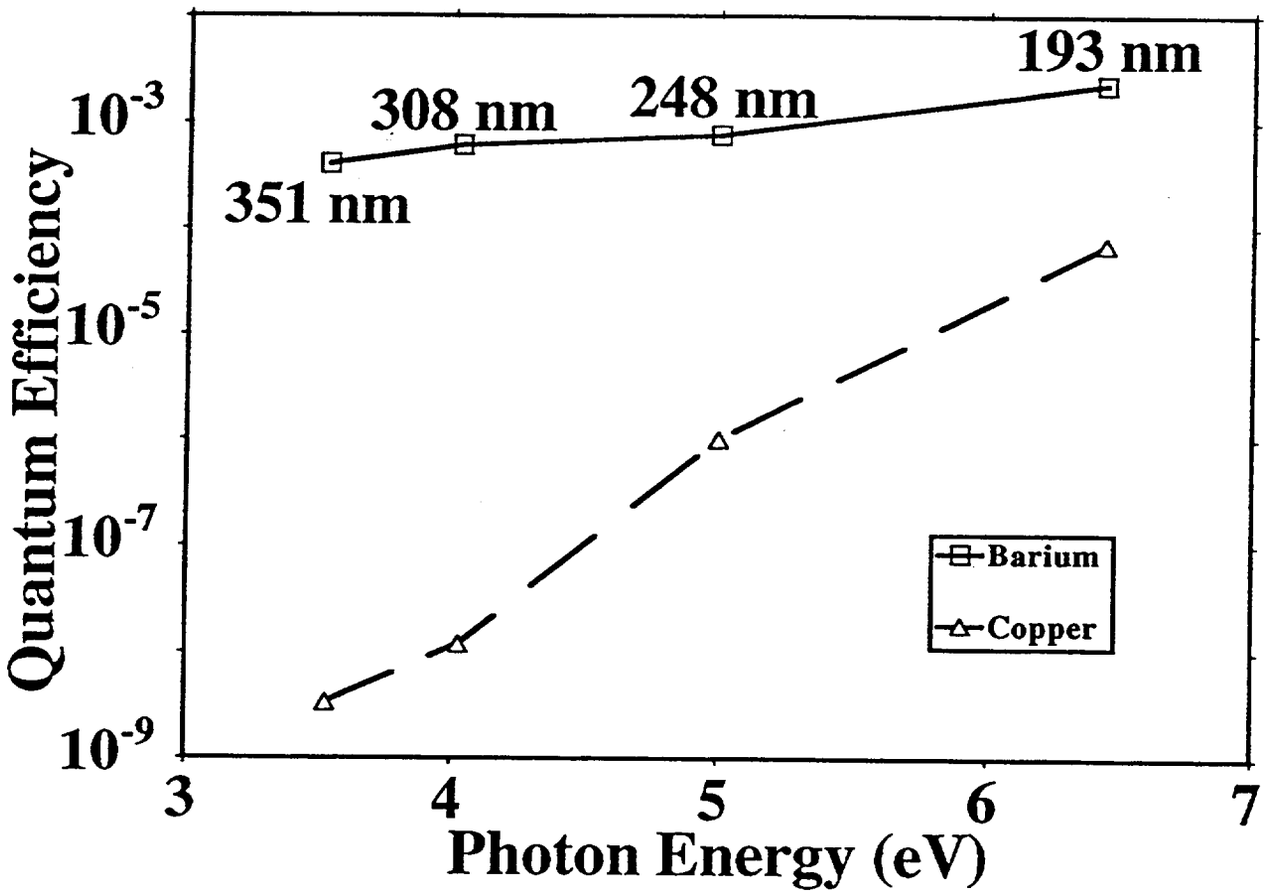
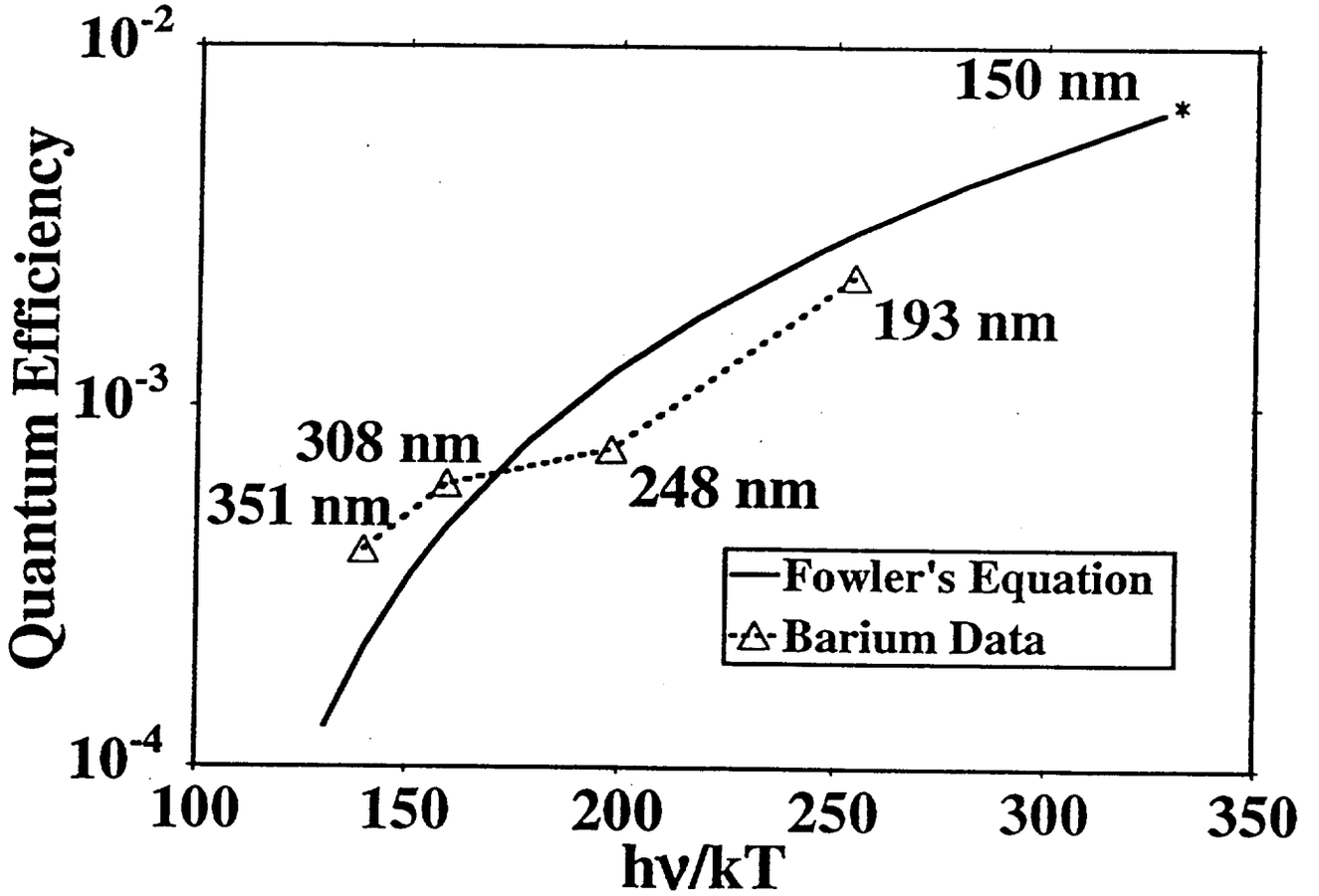

193 nm
308 nm
248 nm
351 nm
10^{-3}
10^{-5}
10^{-7}
10^{-9}
Quantum Efficiency
Barium
Copper
3
4
5
6
7
Photon Energy (eV)


Quantum Efficiency
10^{-2}
10^{-3}
10^{-4}
150 nm
193 nm
308 nm
351 nm
248 nm
Fowler's Equation
Barium Data
100
150
200
250
300
350
hν/kT