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Conversion of heat and light simultaneously using a vacuum photodiode and the thermionic and photoelectric effects[☆]

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Abstract

The photoelectric and thermionic effects are combined in an illustrative experiment to demonstrate that solar light and heat can be converted into electrical energy simultaneously. When an electron is ejected from the cathode and is collected by the anode, a difference in chemical potential develops between the anode and cathode Fermi levels. Work can be extracted as the electron returns to the emitter Fermi level via the load. When the electron is not thermalized, it is said to be a "hot" electron. Ross and co-workers have predicted that the AM1.5 efficiency limit for a hot carrier conversion system, 66%, is greater than that for a purely thermal system, 52%, or for a quantum system, 33% (e.g., a photovoltaic cell). The present work was undertaken to provide an easy to reproduce experimental format to explore these concepts. As an example suitable for a student laboratory, a commercial vacuum phototube is used as a quantum and thermal energy converter. An S1 photocathode comprised of Ag_2O:Cs is employed at low temperatures, $T < 100°C$, to demonstrate that the power converted by a heated and illuminated phototube is greater than that obtained either heated in the dark, or under illumination at room temperature. Although the conversion efficiency and power production is small in this example (approx. 10^{-3}%), the experiment demonstrates how two forms of solar energy can be simultaneously utilized. It also promotes a thermodynamic approach to the evaluation of solar converters. The use of cesiated III/V materials (e.g. InGaAsP:Cs) as photocathodes is discussed as a possible research pathway for realizing efficient hot electron devices.

Keywords: Vacuum phototube; Thermionic emission; Ag_2O:Cs; Hot electron; Education

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1. Introduction: phototube photodiodes and the photoelectric effect

In 1887, long before the discovery of modern photovoltaic (PV) solar cells and detectors, it was discovered that the irradiation of some metals could result in the ejection of electrons [1]. This "Photoelectric Effect" produces a voltage and current when light of the proper wavelength shines on a cleaned surface [2]. For most metals, the wavelengths of light that stimulate this effect are in the ultraviolet region, and the photocurrents are feeble. Later, it was found that semiconductors such as CsSb, Ag_2O:Cs (cesiated silver oxide), Si, GaAs and InGaAs exhibit the photoelectric effect, with the III/V materials exhibiting the highest quantum efficiencies (electrons emitted per input photon). These materials can detect visible, and even near infrared wavelengths, and are employed as sensitive and fast time response photodetectors and photomultipliers [3].

The conversion of light into electrical energy using the photoelectric effect has been previously demonstrated [1]. This effect has been suggested as a possible converter of solar energy [4]. Likewise, the thermionic effect, where electrons are ejected by heat instead of light, has been suggested as a converter of waste heat in nuclear reactors or in solar conversion systems [5,6]. In this paper, both these effects are combined in a simple experiment to show that heat and light can be converted into electrical energy simultaneously. In contrast to traditional photovoltaics (PVs), this is possible because the light absorber can be maintained at a higher effective temperature than the electron collector.

If the electron is ejected from the cathode, and is collected by the anode, a difference in chemical potential develops between the Fermi levels of the anode and cathode. Work can be extracted as the electron returns to the emitter Fermi level via the load. In contrast to a standard PV solar cell, the electron is not necessarily thermalized to the lower level of the excited band before it is collected. When the electron is not thermalized, it is said to be a "hot" electron. Since excess entropy is not generated by thermalization, a phototube photodiode has been employed to measure the spectral distribution of a light source without the use of a monochromator by simply varying the applied voltage [7]. Since entropy generation is less than for a PV cell, a hot electron converter could, in principle, exceed the efficiency limits for standard solar cells.

Ross and co-workers have previously proposed the collection of hot electrons for solar energy conversion, and have developed a theoretical model based on a thermodynamic approach to efficiency limits [8]. This work has shown that the ideal product of the electron charge and maximum voltage in the hot electron converter is not merely equal to the chemical potential of the excited states in the absorber. This is unlike the case for a standard PV solar cell. Further, it was shown that the AM 1.5 efficiency limit for the hot carrier collection conversion system can be greater than that for a purely thermal system or for a quantum system (e.g. a PV cell) [8]. This has prompted solar researchers to search for, and develop, practical hot carrier solar conversion systems.

Photoelectron emission has been observed in aqueous solutions of CdS and ZnS colloidal particles [9], and in GaP photoelectrochemical cells [10]. More recently, Martin Green and co-workers at the University of New South Wales have proposed hot electron conversion as part of an effort towards third generation PVs for high conversion efficiency at low cost [11]. Other researchers have demonstrated a PV device resembling a dye sensitized solar cell that is based on internal electron emission [12]. These studies have stimulated the present work whose goal is to show how hot electron converters can be easily demonstrated in an educational setting.

Presented in this paper is an experimental set-up, suitable for a student laboratory, in which a phototube is used as a hot electron energy converter. An S1 photocathode comprised of Ag_2O:Cs is employed at low temperatures, $T < 100°C$, to demonstrate that the power converted by a heated and illuminated phototube is greater than that obtained either heated in the dark, or under illumination at room temperature. Before presenting the experiment, a brief review of the theoretical aspects of this type of conversion is presented to impart a deeper appreciation for the basic underlying physics of this approach. Sections 1–4 are suitable to give to graduate or undergraduate students, while Section 5 is provided for those who wish to delve further into combined energy conversion.

2. Photoconversion and thermal conversion with phototubes

2.1. Basic theory of photoemission

The upper portion of Fig. 1 shows the relevant standard energy band diagram, together with the energy distribution of the electrons in the photocathode, $n(E)$, and the collected electrons, $N(E)$ [13]. These would, for example, be in units of $\mathrm{cm}^{-3}\,\mathrm{eV}^{-1}$. A similar diagram would describe the case using a degenerate, or metal-like, semiconductor cathode. In a photodiode, only a fraction of the emitted electrons are collected, so $n(E) \neq N(E)$. The fraction collected depends on the voltage, V, the work function of the materials and input photon energy, e. Following standard semiconductor physics terminology, as has been utilized above, the density of states is given by $D(E)$, while φ_a and φ_c are the anode and cathode work functions, respectively (refer to Fig. 1). The Fermi levels are denoted by E_f, where E is the energy of electrons relative to the bottom of the band. For a simple metal photoemitter, light excites an electron to the upper level of the cathode (or emitter) valence band (VB) where it may be emitted if it possesses sufficient energy to escape [13]. For a semiconductor cathode, the electron must be elevated to a level in the conduction band (CB), but the basic processes are the same. In some photoemitters, a cesium surface layer may also be employed to facilitate electron ejection at the emitter surface [3]. This layer will be discussed later.

It was Einstein who first proposed that the kinetic energy (KE) of the emitted electron is given by the difference between the energy of the incident photon, e, and the work function, φ_c, of the absorber material [1], or

$$\mathrm{KE} = e - \varphi_c. \tag{1}$$

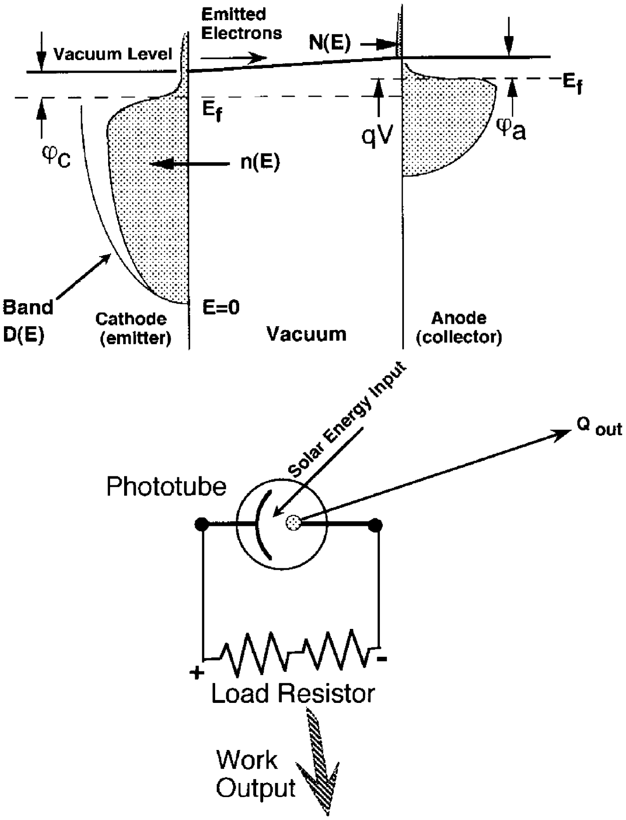


Fig. 1. Top: energy band diagram for a metal-based phototube photodiode showing the electron emission and collection. The anode and cathode work functions, φ are indicated, as well as their Fermi energies, E_f. The quantity qV is the energy associated with the voltage between the two electrodes. The shaded regions are the electron distributions for the cathode, $n(E)$, and for the collected electrons, $N(E)$. Bottom: Schematic diagram for the photocathode and metal anode operated as an energy converter. This is oriented so that it corresponds to the band diagram.

At the so-called saturation potential, all the electrons which posses sufficient energies to leave the emitter also possess kinetic energies that allow them to reach the collector. This scenario is analogous to the short circuit current condition for a silicon solar cell. Most phototube detectors are operated under these bias conditions. In this case, the device consumes energy from an external supply. If, however, energy is to be generated by a photodiode operating as a solar converter, a voltage must develop such that the Fermi level of the collector rises above that of the emitter as is shown in Fig. 1. A solar converter must also produce a net output photocurrent.

2.2. Net photocurrent

The output current density (current per unit area) is given by the symbol J, and the voltage of the converter by V. Note that the term V given here is a voltage and not a velocity. In the convention followed by this paper, positive J values represent generated (output) currents. One will note in the experimental section that these are produced at negative bias voltages. If the current density from the cathode (emitter) is J_c, and the anode current density is J_a, then net current, J, [5] from the photodiode is given by

$$J = J_c(V) - J_a(V). \tag{2}$$

The anode current is produced from any stray light that is incident on the anode, in addition to the thermal excitation of carriers at the temperature of the anode. If the electron is collected by the anode, it can then be thermalized to the anode Fermi level before producing work. This will produce heat (Q_{out}) that will need to be removed from the conversion system (refer to Fig. 1).

If the anode and cathode are maintained at different temperatures, a phototube photodiode can convert heat into electricity. If it is simultaneously illuminated, this light energy can also be converted into electricity. This property of simultaneous conversion is verified by the experimental work presented in the following section. The set-up can be easily reproduced, modified and explored in a student-oriented format. The experiment is thus useful for illustrative and educational purposes, and it is presented for these purposes in this work, rather than suggesting that it be used as a practical solar converter.

3. Phototube experimental set-up

The experiment follows from one described in the historical literature [14]. A modern commercially available phototube and relevant solid-state amplifier techniques are employed in the measurement of the voltage, V, as has previously been discussed [15–17]. The illuminated photocathode of a vacuum photodiode (Hamamatsu PV11) was heated with a resistor, or in a water bath, to a temperature less than 100°C (see Fig. 2). The cathode is on the inner surface of the glass envelope, and the anode is a sphere inside the tube. A temperature difference develops between the anode and cathode when a $68\,\Omega$ resistor, which was pressed on the outside of the phototube heats the glass envelope. An input thermal power of 2.38 W was supplied by this resistor, which acted as a heater when connected to an external power supply (3–12 V). An input optical power of 4–$10\,\mu\mathrm{W/cm^2}$, as measured by a calibrated Silicon photodetector, was supplied to the phototube from a Hewlett Packard HLMP 8104 red light emitting diode (LED) (650 nm). The use of the collimated beam from the LED allowed for the illumination of a $1\,\mathrm{cm^2}$ area of the cathode while maintaining the anode in the dark. In this case, the raw current values, I, are numerically equal to the current density values, J.

Heating and illumination levels were carefully adjusted so that the current output of the phototube due to each was comparable. This necessitated lowering the LED light output so that the photoelectric response (to the LED) was close to the thermionic response when the cathode was heated in the dark. The current–voltage and power generation characteristics were obtained using a Keithley model 616

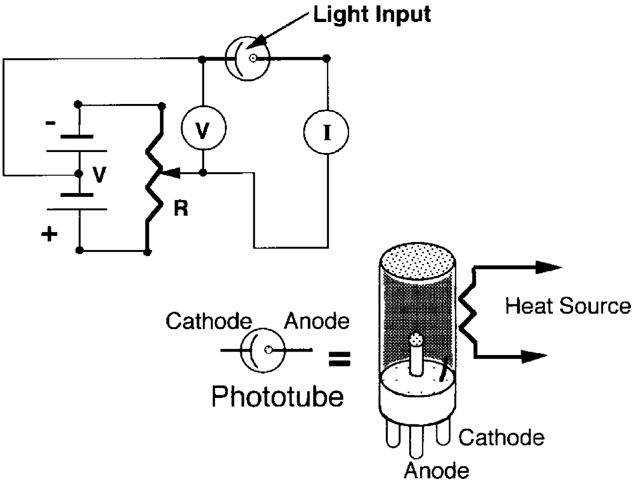


Fig. 2. Schematic circuit diagram used to determine the current–voltage ($I-V$) and the corresponding power output characteristics of a commercial Hamamatsu PV11 photodiode. The photocathode of the phototube (the electron emitter) is deposited on the inner surface of the transparent glass envelope and is heated by a resistor attached to the tube and connected to an external power supply. The anode is a metal sphere suspended inside the tube. Light input is from a collimated red LED. A variable resistor allows one to scan the full $I-V$ curve.

electrometer and Hewlett Packard E2378A voltmeter (see schematic in Fig. 2). Data was taken either by point to point manual measurements or with the aid of a data acquisition system and computer.

Care was taken to avoid: (1) heating the phototube to high temperatures ($T > 100°\mathrm{C}$), and (2) application of excessive bias voltages ($V > 5\,\mathrm{V}$), as these may cause evaporation of the tube's photosensitive coating. The exact equipment and materials for the experiment are not critical as long as the basic intent, as described above, is followed. The experiment lends itself to student improvisation and optimization.

4. Phototube experimental results

For orientation purposes, the full $I-V$ curve measured using the set-up of Fig. 2 is shown in Fig. 3A for an irradiance of $6\,\mathrm{mW/cm^2}$. Under the sign convention used in this work, only the region of negative voltages generates power. Fig. 3B shows the corresponding energy diagrams for two points labeled a and b. These band diagrams illustrate the case for a metal cathode or, with minor modifications, for the case of a degenerate (metal-like) semiconductor photocathode. The saturation condition corresponds to point "a" and the stopping potential is near point "b". The exact locations are difficult to define at temperatures above absolute zero due to the "tail" in the electron distributions $n(E,e)$ and $N(E,e)$ that extends to higher electron energies (see Figs. 1 and 3B). It is exactly this "tail" and ambiguity that results in the ability to convert heat and light energy into work simultaneously. At absolute zero temperature, the maximum photoelectron energy is collected at the stopping potential point [1,17]. At temperatures above absolute zero, the electrons are present in the "tail" of the electron distributions, and some of these can also be collected.

To produce an external electrical energy output, the emitter (photocathode) need only be maintained at temperatures higher than the collector (anode), or the photocathode can be illuminated [6]. The results of current–voltage measurements undertaken using this concept and the apparatus of Fig. 2 are shown in Fig. 4A. One should note that in experiments where both the anode and cathode electrodes were allowed to reach the same temperature, the synergistic effect between heat and light disappears as is expected from Eq. (2).

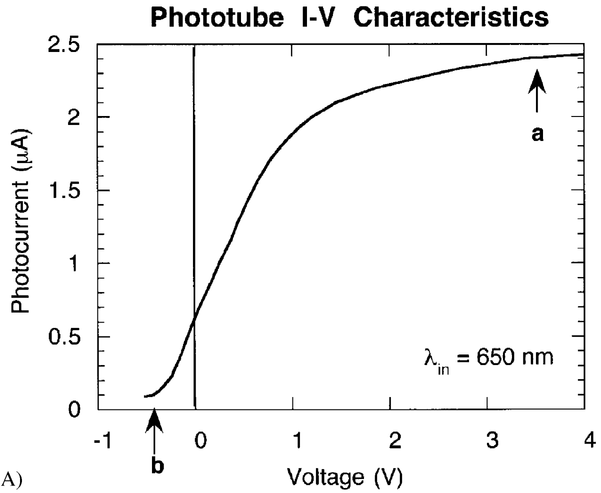
From the power production curve of Fig. 4B, one sees that power can be produced, in a synergistic manner, by the simultaneous action of heat and light on the photocathode. This is in contrast to a PV or photochemical cell. For a standard PV cell, an increase in temperature may increase the photocurrent, via the kinetics or mobility of the material, but it will necessarily lower the photoconverter's voltages [18]. Given the input power to the phototube converter, the energy conversion efficiency in this experiment is admittedly small, and is on the order of $10^{-3}\%$, but the experiment satisfies the goal of demonstrating the synergistic effect.

5. Discussion and future work

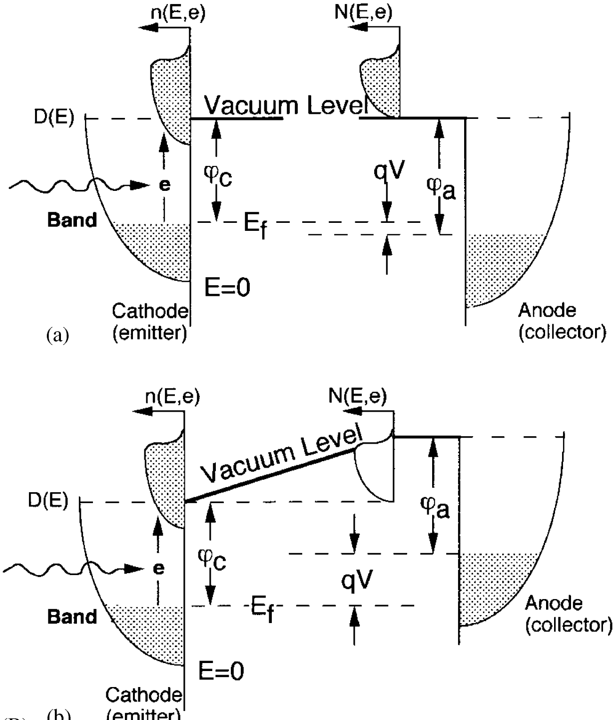
As illustrated in the experiment, heat and light can be simultaneously converted to work using a vacuum photodiode. This is possible because the photoelectric and thermionic effects allow for different cathode and anode temperatures in the converter [4–6]. Although the conversion efficiency and power production is small in the present example, this work demonstrates, in a easy to reproduce experiment, how two forms of energy can be simultaneously utilized.

5.1. Advanced topic: hot carrier converter thermodynamics

For those students who would like to go beyond a basic understanding, the following section outlines why heat and light can, at least in principle, be converted at high efficiencies. Ross and co-workers have developed a theoretical framework for calculating the efficiency of a hot-carrier solar converter based on detailed balance and thermodynamics [8]. It stems from the observation that the recombination in a solar cell is either radiative (producing light), or non-radiative (producing only lattice vibrations or heat). Ross postulated that the radiative portion can be described by the generalized Plank equation [8,18] modified to include hot electron effects. Although it may seem counterintuitive at first, the emission of light from a photoemitter is well documented. It is also known that an "inverse photoemission" effect may result in the production of light from electrons incident on a material in a vacuum [19]. Likewise, spontaneous luminescence from hot electrons has been used to measure the electron effective temperature [20]. In addition, researchers have reported anti-Stokes photoluminescene (photon energy up-conversion) from size-quantized semiconductor colloids [21]. Like a traditional PV solar cell, a phototube photodiode therefore can, and does, emit luminescent radiation. Typically,



(A)



(B)

non-radiative recombination is so strong as to make luminescent (radiative recombination) almost undetectable [18]. In a more efficient solar converter, it is desired that all forms of recombination are minimized.

The significant findings of Ross and co-workers include the idea that a solar conversion system that collects hot electrons results in an absorber with a chemical potential, μ_{H} at a effective temperature of the hot (non-thermalized) electrons, T_{H} [8]. Ross has also shown that the ideal output current density, J, and voltage, V, are a function of the input spectrum, and output radiative recombination (luminescence). This approach is outlined below.

In the absence of non-radiative recombination, the ideal, or limiting, cathode current density, J_{c}, is therefore given [8] by the difference between the excitation current, J_{SC}, and radiative recombination current, J_{r}, or

$$J_{\mathrm{c}}(V) = q \cdot \int N(E, e)\,\mathrm{d}E = J_{\mathrm{sc}}(e) - J_{\mathrm{r}}(\mu_{\mathrm{H}}). \tag{3}$$

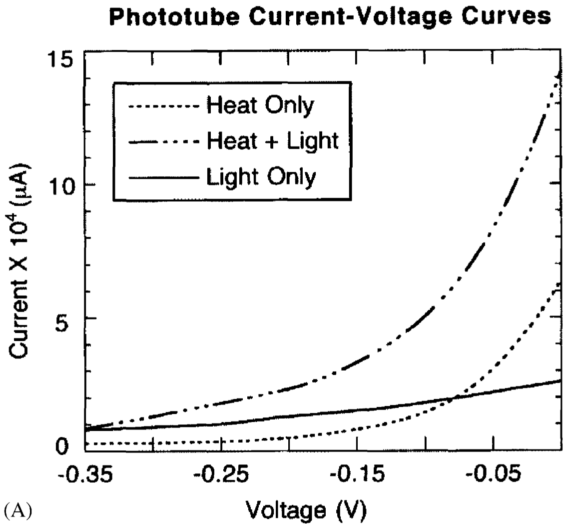
The terms in the parentheses indicate that the symbol preceding them is a function of those quantities. The quantity q is the electron charge, and the chemical potential is given by μ_{H} at an effective temperature of T_{H}. The maximum current that can be produced by the converter is limited to the number of solar-excited charge carriers produced per unit area, J_{SC} [18]. In the case of solar illumination, the generated current, J_{SC}, is given by

$$J_{sc} = q \int n(E, e)\,\mathrm{d}E = q \int a(e)\Gamma(e)\,\mathrm{d}e, \tag{4}$$

where $a(e)$ is the quantum absorptivity [18] of the device, and $\Gamma(e)$ is the input solar photon flux at each photon energy, e. In an ideal device, one producing maximum efficiency, the only form of recombination is radiative, and J_{r} can be calculated from the generalized Planck equation [8,18].

The first integrals of the above equations, involving $N(E, e)$ and $n(E, e)$, may be solved by a complex approximation of Fermi–Dirac-based equations. This yields the Fowler–Nordheim and Richardson–Laue–Dushman equations [22]. Fortunately, the alternative approach of Ross represented by the last portions of Eqs. (3) and (4) use Bose–Einstein statistics and the generalized Planck equation to accomplish the same task. This approach results in simplifications from which one gains useful insights into the efficiency limits of hot electron energy converters [8]. Utilizing the

Fig. 3. (A) Typical room temperature current–voltage characteristics for the Hamamatsu PV11 photodiode illuminated (at 6 mW/cm^2) by a red LED. Point "a" corresponds to saturation, where most photomultiplier tubes are operated, and point "b" is near the "open circuit" voltage (e.g. stopping potential). Negative voltages indicate operation as a power generator (net power production to the load), while positive voltages indicate operation as a biased photodetector. (B) Schematic energy band diagram for two points in the current–voltage curve of (A). The cathode and anode can be subjected to a difference in temperature, and one electrode can be illuminated (wavy line) by a photon of energy, e. Photoelectrons as well as thermionic electrons then contribute to the net current. Shown in the top diagram (labeled "a") is the case for the saturation current point. Shown at the bottom (labeled "b") is the position of the energy levels near the stopping voltage. The shaded electron distributions (occupancies), $n(E, e)$ and $N(E, e)$, are also indicated, but are rotated by 90° from their typical orientation (y and x axes are interchanged).



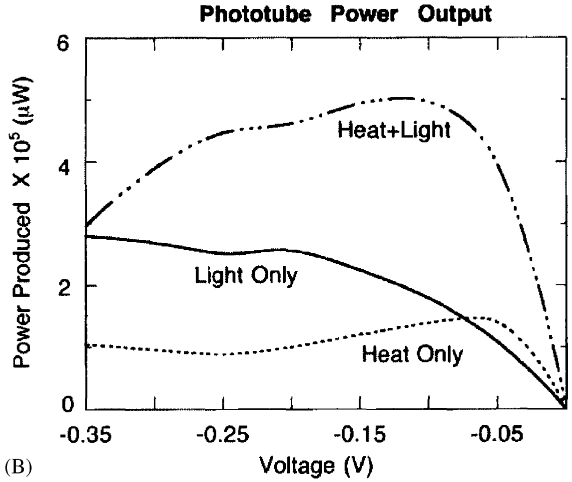


Fig. 4. (A) Current–voltage characteristic of a PV11 phototube in the power production region of the curve. Dash-dotted line: cathode irradiated with an LED at 4 μW/cm^2, and heated at the same time. The irradiated area was 1 cm^2, solid line: cathode irradiated with an LED, at 4 μW/cm^2, but at ambient temperature. Dotted line: cathode heated to a temperature of 100°C, but with no irradiation. In all cases, the anode was at ambient temperature in the dark. (B) Power produced by a phototube corresponding to the $I-V$ curve of (A). The power converted by a heated and illuminated phototube is greater than that obtained either heated in the dark, or under illumination at room temperature.

generalized Planck equation, it was found that J_c increases as the effective temperature of the absorber increases, or as the incident light intensity increases [8]. This thermodynamic approach therefore illustrates that the photoelectric and thermionic effects could, in principle, allow for synergistic effects during the conversion of heat and light.

Ross also equated the effective temperature, ambient temperature, entropy generation and chemical potentials to determine the expected voltage from such an ideal hot electron converter. The chemical potential, μ, as measured at ambient (e.g. 300 K) temperature is given by

$$\mu = \mu_{\mathrm{H}}(T/T_{\mathrm{H}}) + qV(1 - T/T_{\mathrm{H}}), \tag{5}$$

where V is the output voltage of the hot electron solar converter. Unlike a traditional PV cell [18] (where $T = T_{\mathrm{H}}$), the maximum attainable electrochemical potential energy (qV) produced by the hot carrier converter is not equal to the chemical potential of the excited states in the absorber. Because the energy lost to luminescence can be suppressed by negative effective chemical potentials, μ_{H}, in Eq. (5), the AM1.5 efficiency limit for the hot carrier collection system was found to be 66%, compared to 52% for a purely thermal system (e.g. solar-driven Carnot engine), or 33% for a quantum system (e.g. a PV solar cell) [8].

There are several issues for further work suggested by the experiment. Ross has suggested using a negative affinity semiconductor photocathode to approach the higher efficiency limits of hot carrier solar converters [8]. The band diagram for this case is shown schematically in Fig. 5. Shaded areas in this figure are the filled energy levels of the components. Also indicated in Fig. 5 is the electron affinity, χ_{bulk}, in the bulk of the semiconductor, and its bandgap energy, e_{g}. If one applies a monolayer of a low work function material such as cesium on the surface of a p-type

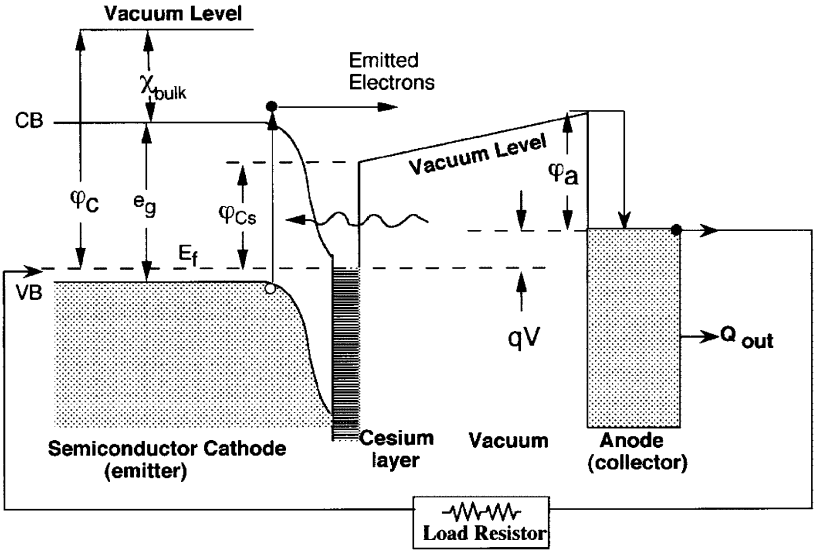


Fig. 5. Energy band diagram for the specific example of a cesiated p-type semiconductor photocathode and metal anode. VB and CB are the valence and conduction bands, respectively. The electron affinity in the bulk of the semiconductor, and its bandgap are indicated. The emitted electron energy is above the vacuum level between the cathode and anode, and the surface is thus said to have a negative electron affinity. This photocathode configuration is extensively used in modern photodetectors, but could also be used as a high efficiency hot carrier solar converter.

semiconductor, band bending, as shown in the diagram can result in the ability of excited electrons to easily be emitted into the vacuum. These photoelectrons tunnel through the depletion region from the CB. As can be seen in Fig. 5, the emitted electron energy is above the vacuum level between the cathode and anode, and the surface of the semiconductor is thus said to have a negative electron affinity. The work function, φ, of Cs is approximately 2 eV [1], and the application of a surface layer of a low work function material like Cs is necessary to produce this negative electron affinity surface condition. Cesiated III/V-based detectors are well known, and exhibit high quantum efficiencies (10–80%) when used as photocathode emitters [3]. Nearly all the electrons excited by light from the VB to the CB can be emitted. The use of p-type cesiated III/V materials (e.g. InGaAsP:Cs) photocathodes, and the engineering of the surface properties of photocathodes, may therefore provide a research pathway for realizing practical and efficient hot electron solar converters.

Replacing the vacuum with an electrolyte and redox couple has also been suggested as a pathway towards practical devices [10,23,24]. In this case, a properly selected redox couple can serve as an energy selective contact to the hot electron emitter. An issue for both vacuum and electrolyte based systems is charge neutralization between electrodes to prevent a space charge that would deflect electrons back to the cathode (origin). Traditionally, this has been accomplished by the addition of Cs ions to vacuum photodiodes [5,6]. Here, the comparison between the dye sensitized solar cell [12,18] may be useful. As seen in the dye sensitized solar cells, and in photosynthesis, an energy barrier is necessary to maintain directional electron flow, to maintain fast kinetics, and to suppress the back reaction. This barrier is present in the phototube converter (see Figs. 1, 3B and 5) and needs to be carefully designed for hot-carrier converters.

There are several embodiments of the ideas presented in this paper. One is the negative electron affinity device. Alternatively, traditional "metallic" or degenerate semiconductors can serve as the photoemitter. It is not clear whether the Ag_2O:Cs photocathode used in this experiment is best described as a metal, degenerate semiconductor, or as a negative electron affinity emitter. Although it is a semiconductor [3], Ag_2O:Cs has a notoriously low quantum efficiency, and was chosen for this work because phototubes utilizing it were readily available. It is therefore suggested, in particular, that the experiment is repeated using newer, and more efficient, photoemitter materials. Geometries and materials that favor field emission [22] should also be considered. Its use in solar concentrator systems could be explored to examine the affect of high $n(E,e)$ concentrations. The simplified experiment presented in this paper should be repeated using these approaches to determine the full potential of hot electron converters.

6. Conclusions

The conversion of light into electrical energy using the photoelectric effect has been previously demonstrated. This effect has been suggested as a possible converter of solar energy. Likewise, the thermionic effect, where electrons are ejected by heat instead of light, has been suggested as a converter of waste heat in nuclear reactors or in solar conversion systems. In this work, both these effects were combined to show that heat and light can be converted into electrical energy simultaneously. In contrast to traditional PVs, this is possible because the light absorber can be maintained at a higher effective temperature than the electron collector. Although the conversion efficiencies in the present work were below $10^{-3}\%$, this system serves as an experimental model to demonstrate conversion in this fashion. An S1 photocathode comprised of Ag_2O:Cs can be used at low temperatures, $T < 100°C$, to demonstrate that the power converted by a heated and illuminated phototube is greater than that obtained either in the dark or under illumination at room temperature. This experiment is easily repeated using more efficient photoemitter materials, and can be modified in a classroom setting to promote further thinking and research in the area of hot electron and combined energy conversion.

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