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THE ULTIMATE EFFICIENCY OF PHOTOSENSITIVE SYSTEMS

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NOMENCLATURE

a	Temperature-dependent coefficient for the frequency distribution of the solar flux at 1 AU, eV/m^2. $\alpha = \gamma \cdot A$.
A	Temperature-dependent coefficient for the frequency distribution of the flux from a blackbody radiator, eV/m^2. $A=2\pi(kT)^3/(hc)^2$.
E_0	Photon energy threshold, eV
f	Frequency distribution for the radiation flux at the receiver, eV/m^2
f_i	Frequency distribution for the entropy flux at the receiver due to internal irreversible (non-isentropic) processes, eV/m^{2}K
f_Q	Frequency distribution for the energy flux at the receiver due to thermal energy exchange with the ambient, eV/m^2
f_S	Frequency distribution for the entropy flux incident on the receiver, eV/m^{2}K
$f_{S'}$	Frequency distribution for the entropy flux radiated from the receiver, eV/m^{2}K
f_u	Frequency distribution for the energy flux incident on the receiver, eV/m^2
$f_{u'}$	Frequency distribution for the energy flux radiated from the receiver, eV/,m^2
f_W	Frequency distribution for the flux of energy converted to useful work, eV/m^2
F_S	Frequency distribution for the entropy flux from a blackbody source, eV/m^{2}K
F_u	Frequency distribution for the entropy flux from a blackbody source, eV/m^{2}K
h	Planck's constant, eV-s
$I_u(y)$	Integral involved in determining maximum efficiency
$I_F(y)$	Integral involved in determining maximum efficiency
k	Boltzmann's constant, eV/K
Q	Heat transferred from the receiver, eV

R_{EO}	Mean radius of Earth orbit, m
R_S	Mean radius of Sun, m
S	Entropy of blackbody radiator, eV/K
T	Kelvin temperature
T_R	Temperature of the receiver, K
T_S	Temperature of the Sun, K
U	Energy of blackbody radiation, eV
W	Energy as useful work, eV
x	Receiver to source temperature ratio, $x=T_R/T_S$
y	Ratio of threshold energy to source energy, $y=E_0/kT_S$
z	Ratio of photon energy to source energy, $z=h\nu/kT_S$
Δf_F	Change in free energy of the radiation fields, eV
γ	Geometrical factor giving diminution of flux from source to receiver, $\gamma=(R_{EO}/R_S)^2$
η	Solar energy conversion efficiency
η_{max}	Maximum solar energy efficiency for a quantum device
η_0	Limit of maximum efficiency for a threshold device
$\eta(x,y)$	Maximum solar efficiency for a threshold device
$\eta(x,y_0,y)$	Maximum solar efficiency for a bandpass device
$\phi_F(z)$	Dimensionless distribution of free energy flux from a blackbody
$\phi_u(z)$	Dimensionless distribution of energy flux from a blackbody
$\Phi(x,y)$	Free energy available for conversion to useful work by a threshold device
ν	photon frequency, s^{-1}
ν_0	threshold frequency, s^{-1}

The Ultimate Efficiency of Photosensitive Systems

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Summary

Considerable attention is being given to the research and development of systems capable of converting the energy available in the solar flux to a usable form. These efforts cover many different systems including photoelectrochemical (ref. 1), photovoltaic (ref. 2), and biological photosynthetic (ref. 3) systems, each with its own mode of operation. These systems have in common two important but not independent features: (1) they can produce a storable fuel, and (2) they are sensitive only to radiant energy with a characteristic absorption spectrum. General analyses of the conversion efficiencies have been made using the operational characteristics of each particular system (ref. 4). We report here an efficiency analysis of a generalized system consisting of a blackbody source, a radiant energy converter having a threshold energy and operating temperature, and a reservoir. This analysis is based upon the first and second laws of thermodynamics, and leads to a determination of the limiting or ultimate efficiency for any energy conversion system having a characteristic threshold. Results obtained can be easily modified to include the case where the device has a cutoff as well as a threshold. Applications of these results to photosensitive systems of particular interest in recent research efforts will be discussed.

INTRODUCTION

Not all of the radiant energy emitted from a blackbody source at a constant temperature T_S and absorbed by a receiver at a constant temperature T_R can be converted to usable energy; that is, energy in a form other than heat. The basic limitation on the amount of energy converted to a usable form is a consequence of the second law of thermodynamics and it involves the change in free energy of the radiation fields. Some of the radiant energy absorbed by the receiver is converted to heat, some is reradiated, and the remainder is converted by the receiver to another form such as chemical or electrical energy. An upper bound on the efficiency of this conversion, assuming that all of the available free energy was converted to useful work, was derived independently by Petela (ref. 5) and by Landsberg and Mallinson (refs. 6 and 10). This bound is a polynomial function of the temperature ratio $x = T_R/T_S$ given by

$$\eta_{max} = 1 - \frac{4}{3} x + \frac{1}{3} x^4 \quad . \tag{1}$$

Also see reference 7, which provides a comprehensive survey of thermodynamic energy conversion efficiencies.

In real energy conversion devices, not all of the free energy available in the radiation field can be converted to useful work because of basic limitations inherent in the devices themselves; consequently, the upper bound given in equation (1) is too high. One of these limitations affecting the ability of a device to utilize all the free energy available is its spectral response. The spectral response of an energy conversion device is a unique characteristic of that device. In order to insure that our discussion of energy conversion efficiency does not depend on any specific device characteristics, we adopt a simple model for the spectral response. Initially we consider a device with an energy threshold; later we show how the results obtained with this assumption can be used to calculate the efficiency of devices which have an energy cutoff as well. In the analysis that follows, we consider a photosensitive system (or an energy threshold device) to be one in which: (a) no absorbed photon with energy below the threshold value can contribute to useful work and (b) those absorbed photons with energy above the threshold contribute only the threshold energy to the useful work. The energy in excess of the threshold is thermalized to heat energy. Most devices which convert radiation to some other form of energy have a characteristic onset energy threshold. We shall find an expression which gives the maximum efficiency for the conversion of blackbody radiation from a source at temperature T_S to usable energy by a device with an energy threshold E_0 and operating at a temperature T_R in terms of the two dimensionless parameters $x = T_R/T_S$ and $y = E_0/kT_S$.

We emphasize that the only device characteristic appearing in the following calculations is the energy threshold. The approach we have taken follows the concepts developed by Shockley and Queisser (ref. 8) in discussing the ultimate efficiencies of solar photovoltaic devices. We have, however, replaced the specific characteristics of photovoltaic systems on which they base their work by more general thermodynamic arguments. Since we invoke only thermodynamic principles, our results are applicable to any threshold device whether it be physical, chemical, or biological in nature.

ENERGY-ENTROPY BALANCE

First we develop the conditions of energy and entropy balance (ref. 7). Since we intend to describe a system in steady state equilibrium, each thermodynamic quantity is represented by a flux, the amount of that quantity passing through a unit area of the receiver in unit time. Radiation flux will be specified further by a spectral distribution, which is the amount of energy or entropy in frequency interval $d\nu$ centered at frequency ν. Energy and entropy flux distributions F_u and F_S are emitted from a blackbody source at temperature T_S. These are diluted in transit to the flux distributions f_u and f_S which are incident upon the receiver at temperature T_R (see fig. 1). Even without specifying the process occurring at the receiver, we know that the incident flux distribution will be apportioned into various channels as follows. One part appears as reradiated flux distributions f_u' and f_S'. Another part appears as a heat flux f_Q and flux of useful work f_W. These latter quantities represent a rate of energy transport away from the receiver normalized to a unit area of the incident beam intercepted by the receiver. The final quantity f_S^i represents the rate of entropy generation due to processes occurring within the device itself; it is a measure of the energy lost irreversibly in the conversion processes occurring in the receiver. With these definitions of the quantities pictured in figure 1 in mind, we can write out the fundamental balance equations. The energy balance (conservation of energy flux) is given by

$$f_u = f_u' + f_Q + f_W \quad , \tag{2}$$

while entropy balance is

$$f_S = f_S' + \frac{1}{T_R} f_Q - f_S^i \quad . \tag{3}$$

These two equations can be combined to eliminate f_Q and to find the following expression for the flux of useful work

$$f_W = (f_u - T_R f_S) - (f_u' - T_R f_S') - T_R f_S^i \ . \tag{4}$$

In classical thermodynamics, the work done on a body in an isothermal reversible process is equal to its change in free energy ($F \equiv U - TS$). Thus we identify the quantities in parenthesis in equation (4) as the free energy flux distribution of the incident and emergent radiation, $f_F = f_u - T_R f_S$. The quantity $T_R f_S^i$ (the irreversible energy loss) is necessarily non-negative by the second law of thermodynamics; consequently, the useful work available in any device is bounded above by the change in free energy

$$f_W < -\Delta f_F = f_F' - f_F \quad . \tag{5}$$

This expression provides the basis for our estimations of ultimate efficiency of threshold devices.

BLACKBODY FLUX DISTRIBUTION

The equilibrium energy and entropy flux distributions of a blackbody radiator are well known (ref. 9). It is convenient to express these distributions in terms of two universal functions of the dimensionless parameter $z = h\nu/kT_S$. The spectral distributions of the energy and entropy flux emitted from a blackbody at temperature T_S are

$$F_u = A\,\phi_u(z) \quad , \tag{6}$$

$$F_S = \frac{A}{T_S}\left[\phi_u(z) - \phi_F(z)\right] \quad , \tag{7}$$

where

$$\phi_u(z) = z^3/(e^z - 1) \quad , \tag{8}$$

$$\phi_F(z) = z^2\,\ell n(1 - e^{-z}) \ , \tag{9}$$

and

$$A = 2\pi\ (kT_S)^3/(hc)^2 \quad . \tag{10}$$

These flux distributions, which are diluted as they propagate from their source, arrive at the receiver diminished by a geometrical factor γ. If we consider the Sun as the source of the radiation and consider an Earth-based receiver, then this geometrical factor depends on the ratio of the radius of the Sun to that of the Earth's orbit, that is $\gamma = (R_S/Reo)^2$,

$$f_u = a\,\phi_u(z) \tag{11}$$

$$f_S = \frac{a}{T_S}\left(\phi_u(z) - \phi_F(z)\right) \quad , \tag{12}$$

where $a = \gamma A$. The magnitude of the incident flux as given by a is important only in that it depends upon the cube of the source temperature. Using the measured average of the total energy flux incident on the Earth's surface, Landsberg estimates the effective blackbody source temperature to be T_S = 6000 K (ref. 10).

LIMITS TO ENERGY CONVERSION BY THRESHOLD DEVICES

As a first step in discussing the energy conversion processes in a threshold device, we examine the amount of free energy available in the incoming radiation. The incident free energy flux distribution depends upon both the distribution of the incident radiation and the temperature of the receiver. In order to remove the intensity of incoming radiation from the discussion and so treat only universal functions, we consider the ratio Φ of incident free energy utilized to the total incident energy, where

$$\Phi \equiv \int_{\nu_0}^{\infty} \frac{h\nu_0}{h\nu} (f_u - T_R f_s) d\nu \Big/ \left(\int_0^{\infty} f_u d\nu\right) \quad . \tag{13}$$

This expression gives the fraction of free energy available for conversion to useful work by a device with threshold energy $E_0 = h\nu_0$. The function Φ depends only on x and y defined previously

$$\Phi\ (x,\ y) = \frac{15}{\pi^4} \cdot y \cdot \{(1 - x)\ I_u(y) + x\ I_F(y)\} \quad , \tag{14}$$

where

$$I_u(y) = \int_y^{\infty} s^2 (e^s - 1)^{-1}\ ds \quad , \tag{15}$$

and

$$I_F(y) = \int_y^{\infty} s\ \ell n\ (1 - e^{-s})\ ds \quad . \tag{16}$$

Several important characteristics of the energy conversion process can be inferred from the graph of the function $\Phi(x,y)$ shown in figure 2. When the receiver temperature is 0 K, all of the internal energy in the incident radiation is available for conversion; just how much is used by a specific device depends on the value of the threshold parameter $y = (h\nu_0/kT_S)$. This curve (the uppermost curve in fig. 2) was first obtained in reference 8 and has a maximum of about 0.44 at $y = 2.2$. As the temperature of the receiver increases, the energy available at all threshold values decreases as the maximum shifts to higher values of y. When the receiver is at a temperature greater than 0 K, it acts as a thermal barrier requiring that some of the incident energy is transformed to heat and consequently unavailable for conversion to useful work. The higher the receiver temperature relative to that of the source, the more incident energy is lost in this way. When the value of T_R/T_S reaches about 0.7, almost all of the incident energy is converted to heat. For values of the temperature ratio larger than 0.7, more energy is required to maintain the receiver temperature than is coming in from the radiation and the receiver draws heat from its surroundings. Thus the importance of the temperature ratio x emerges: the higher x becomes, the more difficult it is to extract energy from the source.

THE ULTIMATE EFFICIENCY OF A THRESHOLD DEVICE

In order to determine the efficiency of our threshold device to conversion of blackbody radiation, we consider a special configuration of source and receiver. Since the efficiency depends only upon the intrinsic characteristics of the device and the radiation, and does not depend upon details of geometry, we evaluate the efficiency in a convenient geometry.

The special configuration we use to determine the ultimate efficiency is shown in part (a) of figure 3. Radiation from a source in equilibrium at temperature T_S falls on a receiver at temperature T_R . We do not specify how the receiver maintains the fixed temperature, but it reradiates energy with the characteristic emission from a blackbody at this temperature. We define the efficiency of this device to be the ratio of the change in free energy flux utilized by the device to the total energy flux incident upon it. The incident free energy flux is given by

$$F_F = F_u - T_R \cdot F_S \quad , \tag{17}$$

where F_u and F_S are given by equations (6) through (10). The reradiated free energy flux that is emitted from a blackbody at temperature T_R is

$$F_F' = F_u' - F_S' = A\, y^3\, \Phi_F(x/y) \quad . \tag{18}$$

The change in the free energy flux, as defined in equation (5) then takes on the form

$$\Delta\, (F_F) = A\, \{(1-x)\, \Phi_u(y) + x\Phi_F(y) - x^3\phi_F(y/x)\} \quad . \tag{19}$$

The efficiency then is given by

$$\eta \equiv \int_{\nu_0}^{\infty} \frac{h\nu_0}{h\nu}\, (-\Delta F_F)d\nu \Big/ \int_0^{\infty} F_u d\nu \quad . \tag{20}$$

This last expression reduces to

$$\eta(x,y) = \frac{15}{\pi^4}\, y\, \{(1-x)\, I_u(y) + x\, I_F(y) - x^3 I_F(y/x)\} \quad , \tag{21}$$

where the integrals appearing here have been defined following equation (14). Equation (21) gives the efficiency for any threshold device in terms of the two parameters x and y. It is worth noting that, in the limit of non-threshold behavior given by

$$\eta_o = \lim_{y \to o} \eta(x,y)/y \quad , \tag{22}$$

this expression for the efficiency reduces to the polynomial in equation (1). The graph of the family of efficiencies is shown in figure 4. Using this graph the maximum efficiency of several systems of current interest has been determined and the results are displayed in table I.

The generalized energy conversion device that we have discussed here is characterized by a single threshold energy marking the onset of conversion processes. Many devices also have a cutoff energy, and photons with energies above this cutoff do not participate in the conversion process. The maxiumum efficiency function $\eta(x,y)$ defined in equation (21) or its graph in figure 3 can be used to calculate the efficiency of a device with an energy window, a threshold y_0, and a cutoff y_1. A simple argument shows that the maxiumum efficiency of a device with threshold energy $E_0 = y_0 \cdot kT_S$ and a cutoff energy $E_1 = y_1 \cdot kT_S$ is given by

$$\eta\ (x,y_o,y_1) = \eta(x,y_o) - \frac{y_o}{y_1} \cdot \eta(x,y_1) \quad . \tag{23}$$

As an example of this use of the efficiency formula, we consider the cellular absorption spectra for the bacteriochlorophyll a. This spectrum shows a narrow absorption peak in the near infrared; the peak is centered at 883 nm, and has a half-width of 25 nm. Approximating this absorption peak by an energy window with an onset threshold at 858 nm and cutoff at 908 nm, we find an efficiency maximum of 2.4 percent.

While the expression for the maximum efficiency of a threshold device given by equation (21) was derived assuming a special configuration of source and receiver, it describes a general characteristic of the device, and is applicable in other configurations. The configuration for terrestrial applications is depicted in figure 3b, which shows how the flux emitted by the Sun is diminished by a factor of $\gamma \approx 10^{-3}$ before arriving at the receiver. As a result of this diminution, there may not be enough solar energy incident on the receiver to support its reradiation as a blackbody at temperature T_R. If the receiver does reradiate as a blackbody in this case, it must be supplied energy as heat from its surroundings. The magnitude of this contribution of the heat flux does not appear in equation (21) since it was eliminated by combining the equations for energy and entropy flux. In general, the receiver will reradiate with some characteristic emission spectrum necessarily less intense than that of a blackbody at T_R, but regardless of the details of this emission, equation (21) serves as an upper bound on the efficiency.

CONCLUSIONS

Using general thermodynamic arguments we have analyzed the conversion of the energy contained in the radiation from a blackbody to useful work by a photosensitive system. Through this analysis, we have shown that the amount of energy available for conversion is bounded above by the change of free energy in the incident and reradiated radiation field and that this free energy change depends upon the ambient temperature of the receiving device. We have further discussed the limitations imposed upon the receiver due to absorption characteristics and in a general way we have developed a prototypical quantum device to model this absorption. This prototypical device is completely described by two parameters, the ambient temperature of the receiver T_R and the energy threshold of the absorption $h\nu_0$. These two parameters can be normalized by the source temperature T_S to give two dimensionless parameters $x=T_R/T_S$ and $y=h\nu_0/kT_S$. We calculate universal efficiency curves for photosensitive devices in terms of these two parameters, and from these curves determine the maximum thermodynamic efficiency of several solar energy conversion devices currently under study. This analysis indicates that a maximum conversion efficiency of 0.44 is possible for a device with threshold energy ratio of $h\nu_0/kT_S = 2.2$. While an important application of these results is to the conversion of solar energy, the efficiency and availability curves can be applied to the conversion of blackbody radiation in general.

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Table I. Representative threshold devices, their threshold energies, typical operating temperatures, and theoretical measured efficiencies to terrestrial solar radiation.

Converter	Threshold Energy (eV)	Operating Temperature	Theoretical Efficiencies	Measured Efficiencies
Solar Cell Si	1.11	300 K	40%	16%
Liquid Junction Solar Cells InP	1.28	300 K	39%	15%
Thermoelectric Converter (T_S=1000 K)	0.1	300 K	20%	12%
Photo-electric Generator (Cesium)	3.9	300 K	6%	
Thermophotovoltaic (2300 K - Si)	1.11	300 K	15%	-
Photochemical System	2	300 K	27%	-
Biological System Absorption peak of bacterio-chlorophyll a	858 nm to 908 nm	300 K	2.4%	- -

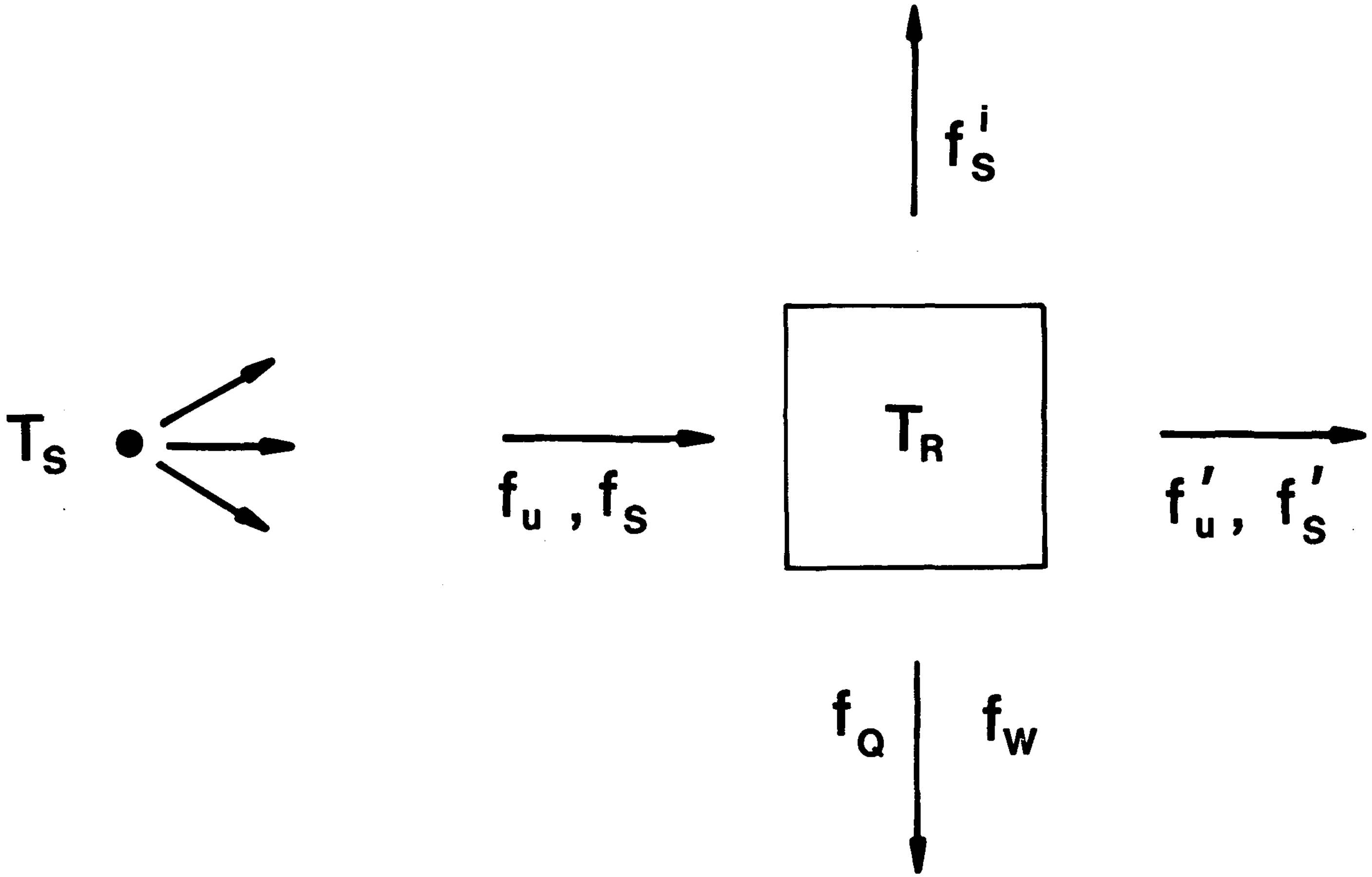

Figure 1. A schematic diagram of the energy and entropy flux distribution emitted from a source at temperature T_S and arriving at a receiver at temperature T_R (f_u, f_s). Part of this flux is reradiated (f_u', f_s'), the remainder appears as heat flux f_Q, usable work f_u, or energy irreversibly lost f_s .

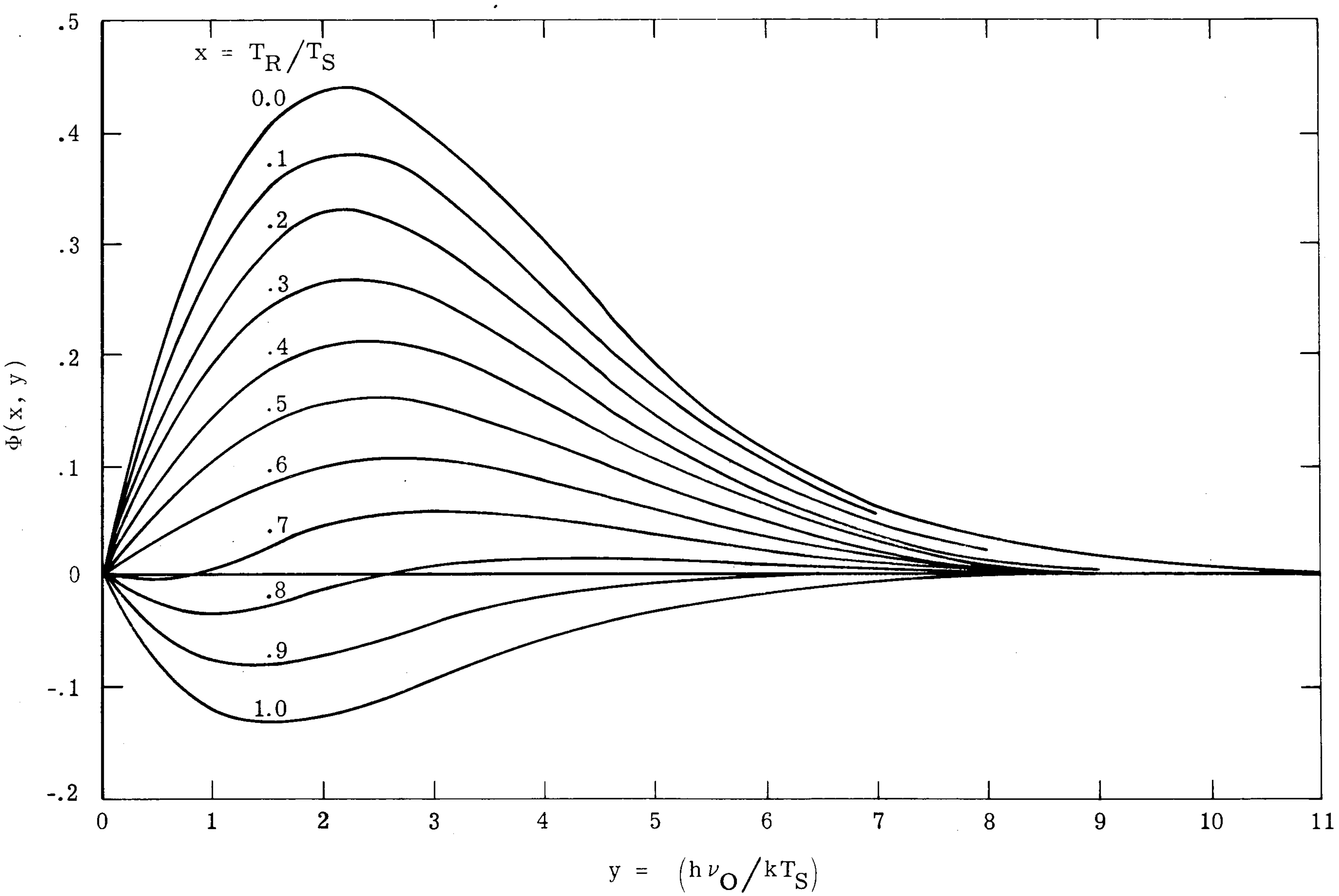

Figure 2. Fraction of total energy from a blackbody source at temperature T_S available for conversion to useful form by a device having an energy threshold $h\nu_O$ and operating at constant temperature T_R, $\phi(x,y)$ expressed in terms of the dimensionless parameters $y = h\nu_O/kT_S$ and $x = T_R/T_S$.

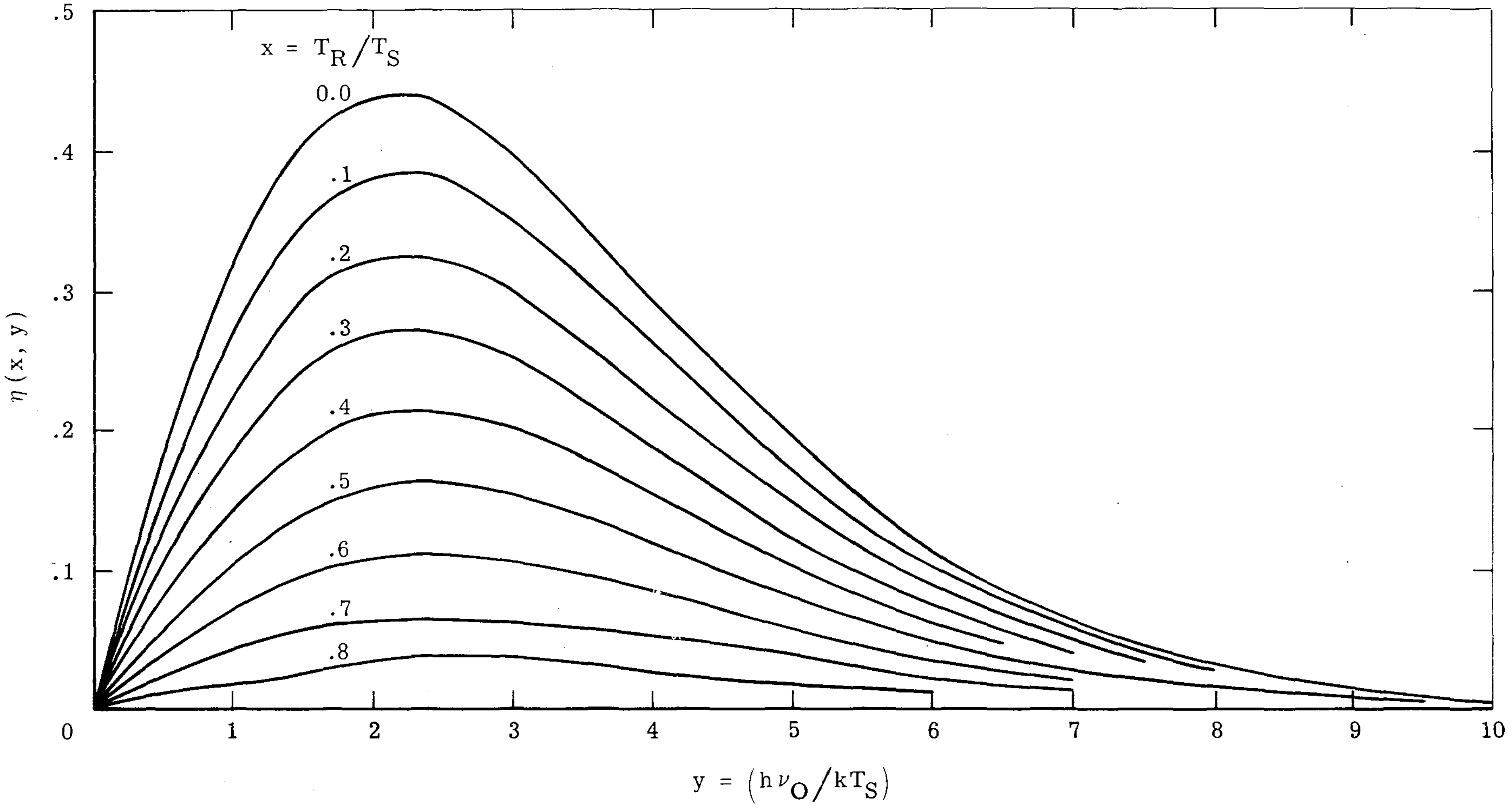

Figure 3. Maximum efficiency for the conversion of energy from a blackbody source at temperature T_S by a device having an energy threshold E_O and maintained at temperature T_R, η_{max}, expressed in terms of the dimensionless parameters $y = E_O/kT_S$ and $x = T_R/T_S$.

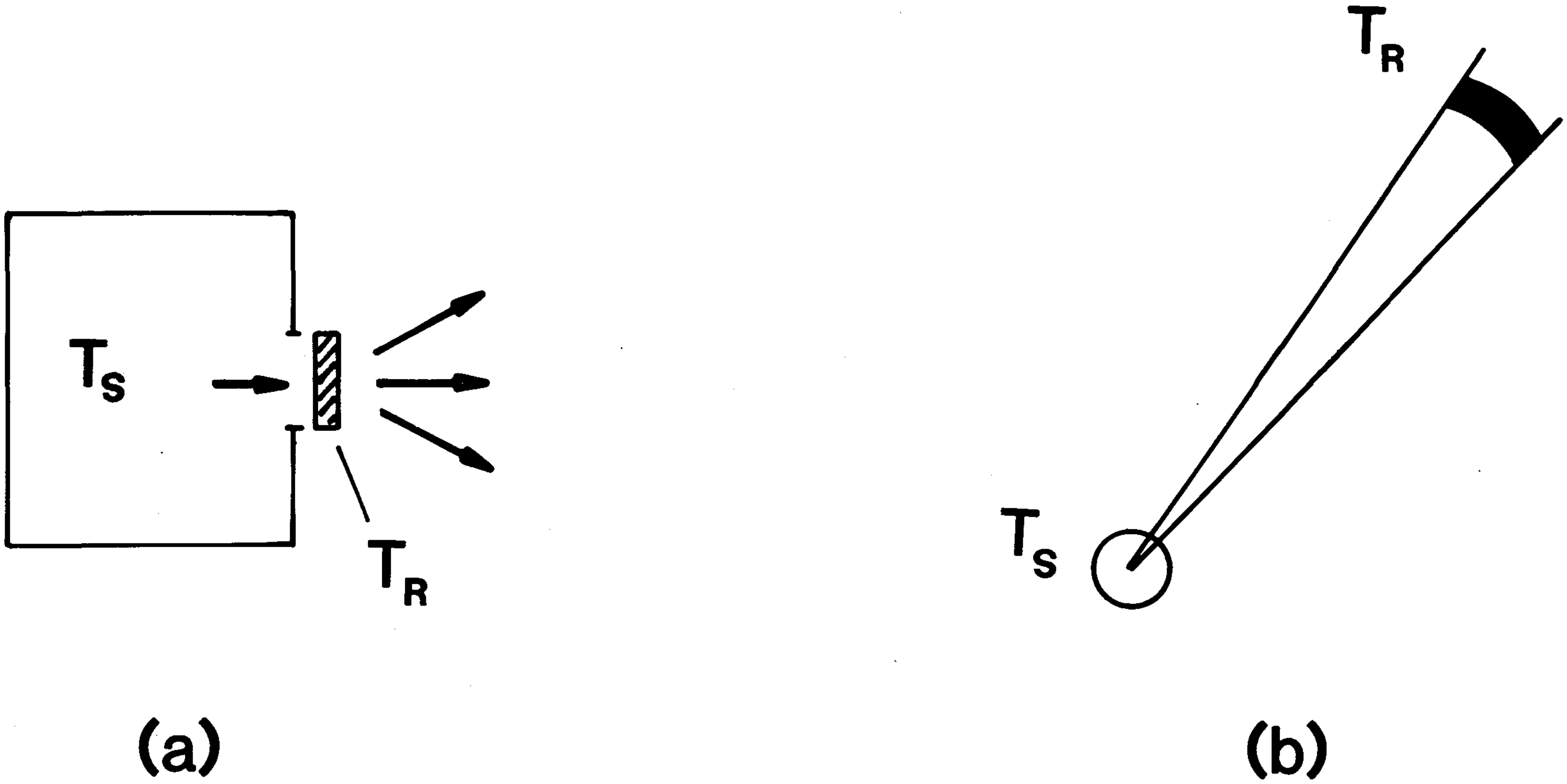

Figure 4. Schematic diagram of the relation between the blackbody radiation source and the receiver: (a) idealized geometry used to find the maximum efficiency of the threshold device, (b) actual geometry for Sun-Earth applications.

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15. Supplementary Notes

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16. Abstract

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