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(Paper No. K-3, 1968 Cryogenic Engineering Conference)

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ABSTRACT

The present paper is concerned with predictions of the total emissivity of metals at cryogenic temperatures based on the anomalous skin effect theory (the ASE theory). An exact numerical result as well as an approximate expression has been developed to represent the spectral emissivity of metals at cryogenic temperatures. The two representations of spectral emissivity after appropriate integrations over wavelength result in, respectively, a closed-form expression and an exact numerical tabulation of total normal emissivity in terms of three dimensionless parameters. These results are then compared with the existing experimental data of the low-temperature total emissivity of metals and with the prediction obtained from the exact numerical integration of the spectral emissivity as given by the Drude single (or free) electron theory (the DSE theory). The comparison reveals the enhanced accuracy of the ASE theory in predicting the radiation properties of metals at cryogenic temperatures.

Introduction

Due to the rapid growth of the cryogenic technology and the accompanying rise in the use of high vacuum insulation, thermal radiation of metals at cryogenic temperatures has become an area of increasing importance. Indeed, a recent review article[1] of the subject indicates the extent to which the development of experimental as well as theoretical techniques involved in this area has rapidly progressed.

Since the primary objective in the consideration of thermal radiation at cryogenic temperatures is the calculation of radiant heat interchange between metallic surfaces, it is only natural that the subject of radiation properties of metals has received much emphasis. It should be noted, however, that the most widely employed basis for the prediction of emissivities of metals has been the Drude single (or free) electron theory of the optical properties of metals.[2] (For the convenience of discussion this theory will be denoted here as the DSE theory.) But in the cryogenic temperature range, the adequacy of the DSE theory is questionable and recourse must be made to what is known as the anomalous skin effect theory (the ASE theory) of the optical properties of metals.[3,4,5] In the region of practical interest for thermal radiation, the predicted emissivity of metals at cryogenic temperatures based on the ASE theory could be one or more orders of magnitude larger than that predicted by the DSE theory.

It is the purpose of the present work to demonstrate the importance of the anomalous skin effect as regards the prediction of radiation properties of metals at cryogenic temperatures. The relative significance of the ASE theory of the optical properties of metals is demonstrated by a comparison of the total emissivity as predicted by both the ASE and the DSE theories. Then

by comparing these predictions with experimentally obtained values of total emissivity, an assessment of the applicability of these theories is carried out.

Theories of Optical Properties of Metals

The region of engineering interest for the calculation of radiation properties of metals at cryogenic temperatures is the far infrared portion of the electromagnetic spectrum. In this frequency range, which lies below the lower frequency limit for internal photoelectric absorption, the absorptivity of the metal can be attributed wholly to the motion of the free electrons. The theoretical prediction of absorptivity requires an analysis of the motion of the free electrons as they interact with the electric field within the metal and as they experience damping due to collisions with the lattice phonons and the defects or impurities. Both the Drude single electron (DSE) theory and the anomalous skin effect (ASE) theory of the optical properties of metals take into account the above-mentioned factors. The major difference in the two theories is that the DSE theory neglects the effects of the space variation of the electric field within the metal whereas the ASE theory takes this spatial variation effect into account.

In the development of the DSE theory, a simple physical model is constructed in which the equation of motion is written for a model electron which represents the behavior of the whole set of electrons playing a role in the absorption process.[2] The damping of the free electron motion is then assumed to be viscous in nature, i.e. with a damping force opposing and proportioned to the velocity of the model electron. The driving electric field is assumed to be a periodically varying function of time only. The solution based on this model can be expressed in the form of the relations between the electrical and optical properties of metals:

$$n^2 - k^2 = 1 - \frac{4\pi Ne^2}{m} \left[\frac{\tau^2}{(\omega^2\tau^2 + 1)}\right] \tag{1a}$$

$$2nk = \frac{4\pi Ne^2}{m} \left[\frac{\tau}{\omega(\omega^2\tau^2 + 1)}\right] \tag{1b}$$

where n is the index of refraction, k is the absorption coefficient, N is the electron number density, m is the electron mass, e is the electron charge, τ is the relaxation time. The relaxation time is related to σ, the d-c electrical conductivity, by the Lorenz-Sommerfeld relation, $\tau = m\sigma/Ne^2$.

As was pointed out previously, the DSE model does not take into account the effect of the spatial variation of the electric field on the free electron motion, i.e. the amplitude of the driving electric field should be a function of the position coordinate. The actual state of affairs within the metal is one in which the amplitude of the electric field decays with distance into the metal. Since the electrons which contribute to the absorption of the electromagnetic energy are those which move within the penetration depth of the decaying electric field, the electrons must necessarily experience some effects due to the spatial variation of this field. Therefore, the DSE theory can be thought of as an approximation which is valid only when the motion (mean free path) of the electrons is small compared to the penetration depth of the electric field. Since the penetration depth is given approximately by $\delta \propto \frac{1}{\sqrt{\sigma\omega}}$ the DSE theory should be applicable when $\sigma\omega$ is small, i.e. high temperatures or low frequency. Under these conditions the electrons "see" an electric field which varies only with time.

In order to predict the optical properties of metals when the penetration depth of the electric field is of the order of the free electron mean free path such as at the conditions of cryogenic temperatures and moderate frequencies,

Reuter and Sondheimer[3] solved simultaneously the Maxwell equations and the Boltzmann transport equation as applied to the free electrons in a semi-infinite medium. The analysis based on the resulting integro-differential equation indicates that the motion of the free electrons in the penetration layer depends upon the electric field everywhere in the metal, i.e. the spatial variation of the electric field. Included in the integro-differential equation is a parameter arising from the boundary condition on the electron motion. This parameter accounts for the nature of the reflection suffered by the electrons at the metal surface. The form of the decaying electric field resulting from the solution of the integro-differential equation is much more complex than the exponentially decaying field resulting from the usual assumption of the current density proportional to the electric field at a given point.

With the specification of the electric field given by the solution of the integro-differential equation, the surface impedance is given by

$$Z = -\frac{4\pi i\omega\mu}{c^2}\,\frac{E(o)}{E'(o)} \tag{2}$$

where μ is the magnetic permeability, c is the speed of light, and E is the electric field, a function of the penetration distance.

For the case of diffuse reflection of the free electrons at the metal surface, which appears to be the prevalent case as indicated by experimental investigation, Dingle[4] obtained an equation for the surface impedance as:

$$\frac{i}{w\bar{Z}} = \frac{1}{\pi}\int_0^\infty \ln\left\{1 + \frac{\xi\, X(t)}{t^2}\right\} dt \tag{3}$$

where $\bar{Z} = (c^2/4\pi\omega\tau\mu v)Z$ (v being the Fermi velocity)

$w = 1 + i\omega\tau$

$\xi = i\,\bar{\sigma}^3\,\bar{\nu}/(1 + i\,\bar{\sigma}\,\bar{\nu})^3$

$$\bar{\sigma} = [v(3\pi m\mu/Ne^2)^{1/2}/c]\sigma$$

$$\bar{\nu} = [2c(3\pi m/Ne^2\mu)^{1/2}/3v]\nu$$

$$X(t) = 2t^{-3}[(1 + t^2)(\tan^{-1} t) - t]$$

Instead of using numerical means to evaluate (3) directly, Dingle obtained two series solutions for the impedance, one in terms of ascending powers of $\xi^{1/2}$, the other in descending powers of $\xi^{1/3}$. The solutions obtained by Dingle are as follows:

for $|\xi| \leq 0.8$

$$\frac{i}{w\bar{Z}} = 1.1547\ \xi^{1/2} - 0.2500\ \xi + 0.1540\ \xi^{3/2} - 0.1262\ \xi^2 + 0.1188\ \xi^{5/2} - 0.1214\ \xi^3 + 0.1307\ \xi^{7/2}\ [-0.148\ \xi^4] + 0.1685\ \xi^{9/2} \cdots \tag{4a}$$

for $|\xi| \geq 0.8$

$$\frac{i}{w\bar{Z}} = 1.1547(\pi\xi)^{1/3} - \{0.1351\ \ln(\pi\xi) + 0.5330\} + 0.0416(\pi\xi)^{-1/3} - 0.0649(\pi\xi)^{-2/3} + (\pi\xi)^{-1}\ \{-0.0164\ \ln\ (\pi\xi) + 0.0184\} + 0.0660\ (\pi\xi)^{-4/3} - 0.0307\ (\pi\xi)^{-5/3} \cdots \tag{4b}$$

The optical and spectral radiation properties can be obtained from the surface impedance given by (4a) and (4b) via the impedance concept.

Relations Between Optical and Spectral Radiation Properties

On the basis of the DSE model, the calculation of the optical properties is quite straightforward. The solution of (1a) and (1b) results in the following:

$$n = \left[\frac{2x}{(y^2 + 4x)^{1/2} - y}\right]^{1/2} \tag{5a}$$

$$k = \left[\frac{(y^2 + 4x)^{1/2} - y}{2}\right]^{1/2} \tag{5b}$$

where

$$x = \left(\frac{4\pi Ne^2}{m}\right)^2 \frac{\tau^2}{4\omega^2(\omega^2\tau^2 + 1)^2} \tag{6a}$$

$$y = 1 - \frac{4\pi Ne^2}{m} \left(\frac{\tau^2}{\omega^2\tau^2 + 1}\right) \tag{6b}$$

By using the complex index of refraction, $\hat{n} = n - ik$, in the Fresnel formulae, the spectral directional reflectivity or absorptivity can be obtained.[2] The spectral emissivity for the case of normal incidence is given by

$$\varepsilon_N(\omega,T) = 1 - \left| \frac{\hat{n} - 1}{\hat{n} + 1} \right|^2 = \frac{4n}{(n + 1)^2 + k^2} \tag{7}$$

This has been calculated using (5a) and (5b) and is shown in Fig. 1 for various temperature levels.

For calculations based on the ASE theory, the analogy between electromagnetic waves and a-c transmission lines is used to obtain the spectral normal emissivity in terms of the surface impedance as follows:

$$\varepsilon_N(\omega,T) = 1 - \left|\frac{(4\pi/c) - Z}{(4\pi/c) + Z}\right|^2 \tag{8}$$

The spectral normal emissivity calculated in this manner using (4a) and (4b) is also shown in Fig. 1.

In the case of the ASE theory, the electric field in the metal is damped in a form other than exponential, so the complex refractive index does not retain the usual physical significance, i.e.

$$E(y) \neq E_o \exp\left[i\left(t - \frac{\hat{n}y}{c}\right)\omega\right]$$

However, it has been shown[5] that if the refractive index is redefined as

$$\hat{n} = \sqrt{\sin^2\phi + \left(\frac{4\pi}{cZ}\right)^2} \tag{9}$$

where ϕ is the angle of incidence, then the Fresnel formulae give the desired spectral directional radiation properties. For cryogenic temperatures and in the far infrared, the following approximation can be made:

$$\hat{n} = \frac{4\pi}{cZ} \tag{10a}$$

or

$$n = \frac{4\pi R}{c(R^2 + X^2)} \quad , \qquad k = \frac{4\pi X}{c(R^2 + X^2)} \tag{10b}$$

where X and R are related through Z = X + iR and can be evaluated from (4a) and (4b).

While the use of the series (4a) and (4b) is satisfactory for machine computations a simple closed form expression would be more desirable from the viewpoint of engineering calculations. Thus, an approximate curve fit was sought for the spectral normal emissivity. A good approximate expression is found to be

$$\varepsilon_N(\bar{\nu},T) = \bar{v}\left(\frac{3}{4} + \frac{\sqrt{3}}{\bar{\sigma}}\right) \{1 - \exp[-\frac{3}{2}(\bar{\nu})^{2/3}]\} \tag{11}$$

where $\bar{v} = v/c$ is the dimensionless Fermi velocity. This expression labeled as the ASE approximation in Fig. 1 has the correct approach to the high frequency limit and agrees quite well with the exact ASE theory in the primary spectral range of the Planck distribution for the given temperatures.

Total Normal Emissivity of Metals

The usual engineering calculation of radiative flux involves the use of the total or integrated hemispherical emissivity. The spectral hemispherical emissivity is related to the spectral normal emissivity as is shown by Dunkle.[6] For the cryogenic temperature range, the spectral range of importance is the far infrared in which the ratio is approximately 1.33 and is relatively independent of wavelength. Thus the ratio of total hemispherical to total normal

emissivity can be also considered as approximately 1.33. In the past much effort has been made to predict the total normal emissivity of metals using the DSE theory.[7,8]

In order to provide a basis for assessment of the applicability and significance of the ASE theory, as opposed to the DSE theory, in predicting the total emissivity of metals, exact numerical computations have been performed using both theories. In the case of the DSE theory, the spectral normal emissivity, given in (7), combined with the optical constants described by (5a) and (5b) was integrated together with the Planck function over frequency to obtain the desired total normal emissivity. Similarly the exact spectral normal emissivity given by the ASE theory was integrated numerically. The results of these numerical calculations are presented in Fig. 2 and Table I, where total normal emissivity can be obtained as a function of the two dimensionless parameters, b_2 and b_3, first developed by Rolling and Tien.[7] These two parameters represent both the temperature and material properties dependence of the total normal emissivity. In the case of the ASE theory, the total normal emissivity depends on the dimensionless Fermi velocity as well as b_2 and b_3.

It is quite evident from the Table I and Fig. 2 that the contribution due to the skin effect can be greater than one order of magnitude. In order to gain a better physical understanding of the total emissivity, calculations were made to obtain total hemispherical emissivity as a function of temperature for copper with various residual resistivity levels (impurity levels). The d-c electrical conductivity was obtained from the Bloch-Grüneisen formula together with Matthiessen's rule.[9] The resulting temperature-dependent total hemispherical emissivities are shown in Fig. 3 for the ASE theory, DSE theory and that based on the Hagen-Rubens relation for the residual resistivities indicated. Also shown are the

existing total emittance data[10,11,12,13] for copper. In addition, the room temperature spectral data of Bennett and Bennett[14] for gold and silver were integrated over the Planck function and are presented in Fig. 3.

In order to provide a simple closed-form engineering approximation to the total normal emissivity as predicted by the ASE theory, the approximate form (11) was integrated together with the Planck function over frequency. This was accomplished by expanding the exponential in equation (11) in a Taylor series expansion and integrating term by term. The result is a series involving Gamma and Zeta functions:

$$\varepsilon_N(T) = \bar{v}\left(\frac{3}{4} + \frac{\sqrt{3}}{\bar{\sigma}}\right)\left[\Gamma\left(\frac{14}{3}\right)\zeta\left(\frac{14}{3}\right)A - \Gamma\left(\frac{16}{3}\right)\zeta\left(\frac{16}{3}\right)\frac{A^2}{2!} + \cdots\right. \tag{12}$$

where

$$A = \frac{3}{2}\left[\frac{2}{3\bar{v}}\left(\frac{3\pi m}{Ne^2\mu}\right)^{1/2}\frac{kT}{h}\right]^{2/3}$$

h is the Planck constant and k is the Boltzmann constant. This series can be approximated by the binomial expansion of

$$\varepsilon_N(T) = 2.47\,\bar{v}\left(\frac{3}{4} + \frac{\sqrt{3}}{\bar{\sigma}}\right)\frac{A}{1 + A} \tag{13}$$

Figure 4 is a comparison of the approximate total normal emissivity (13) with the exact numerical calculations based on the ASE theory.

Discussion

Some insight into the physical significance of the DSE and ASE theories can be gained by a discussion of the spectral absorptivity curves of Fig. 1. First, it should be noted that at 300°K, there is fair agreement between the ASE and DSE theories. This is a consequence of the high degree of damping present at high temperatures which limits the motion of the electrons so that the spatial variation of the electric field has little effect. Second, both

the exact ASE and exact DSE theories approach the Hagen-Rubens relation in the long wavelength limit at both 4.2°K and 300°K. Third, in the near infrared at 4.2°K, the ASE theory predicts a normal absorptivity approximately two orders of magnitude larger than that predicted by the DSE theory. In fact, in this high frequency range, the limiting value of the ASE absorptivity is $\frac{3}{4}\bar{v}$ as seen from the approximate form (11), which is correct for this frequency range. This is the extreme anomalous skin effect region, where the absorptivity is independent of the relaxation or damping phenomena. This is accounted for by the fact that at relatively high frequencies and low temperatures, the free electrons moving within the penetration depth (which is very small compared to the mean free path of the free electrons) experience negligible damping in this very thin region at the surface of the metal. Therefore, in this situation the energy gained by the electrons in the penetration depth is not dissipated immediately. Instead the electrons are driven back into the bulk of the metal by the field where they then interact with the lattice or defects to give up their excess energy in the form of Joule heating. Since the spectral absorptivity is determined by what happens within the penetration depth, this damping, which occurs deep within the metal, does not appear as a parameter in the extreme anomalous skin effect region. The relative difference between the DSE and ASE predictions as exhibited in the spectral absorptivity will, of course, be evident in the total normal emissivity as well.

Figure 2, which allows the calculation of total normal emissivity given the material properties of the metal, illustrates the relative importance of the damping phenomena and skin effect as regards total emissivity. The largest disparity between the ASE and DSE predictions occurs when b_2 is small but b_3 is large. This corresponds to low temperature, high electrical conductivities and

large relaxation time in agreement with our previous discussion of spectral absorptivity. It can also be seen that when the relaxation time, or b_3, is small, the ASE and DSE predictions converge more rapidly with increasing temperatures or increasing b_2.

To provide a specific example for discussion, the total hemispherical emissivity of copper for varying residual resistivity levels are plotted in Fig. 3 as a function of temperature. The total hemispherical emissivity based on both the DSE and Hagen-Rubens formulations show a marked effect due to the varying impurity levels which were introduced via Matthiessen's rule. On the other hand, the ASE prediction was insensitive to the impurity levels introduced. This insensitivity to impurity levels can be accounted for by the fact that those residual resistivities introduced, which become important below about 30°K, are still so small that the resulting differences in the damping are still inconsequential as compared to the skin effect.

Upon comparison of theory and existing experimental data in Fig. 3, it is quite evident that the ASE theory represents a considerable improvement over the DSE theory in the prediction of thermal radiation properties of metals at cryogenic temperatures. The remaining discrepancy between the ASE theory and experimental data is probably due to one of two causes. The real surface effects which are exhibited by the data of Betz et al., Caren, and Fulk and Reynolds would tend to increase emissivity as a result of the increased damping by surface impurities and imperfections. In contrast, the points obtained by integrating the spectral data of Bennett and Bennett are in extremely good agreement with both the ASE and DSE theories at room temperature. The gold and silver samples used by Bennett and Bennett were prepared under extremely stringent conditions of purity and surface finish. It should be noted that

the comparison between measurements performed on gold and silver and predictions for copper is permissible on the basis of the similarity of material properties. Also shown is the data of Ruccia and Hinckley for copper but under less strict conditions of purity and surface finish than those maintained by Bennett and Bennett.

Secondly, there arises the question of the validity of one of the assumptions basic to the formulation of both the DSE and ASE theories, that of the relaxation or damping phenomenon being independent of frequency. Much work has been done recently to obtain the frequency dependence of the damping via a quantum mechanical treatment.[15,16] Inclusion of the effects of frequency dependent damping may improve the predictions still further than has the ASE theory.

Figure 4 shows the extremely accurate agreement of the approximate ASE expression (13) with the exact numerical ASE calculations of total normal emissivity at cryogenic temperatures. Thus, with the use of this simple expression, the total normal emissivity predicted by the ASE theory can be calculated with relative ease.

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Table I. The total normal emissivity as predicted by the ASE theory. (DSE values are in parentheses and the notation -x refers to 10^{-x}.)

b_3	$\bar{v}\times10^3$	$b_2=$ 10^{-6}	10^{-5}	10^{-4}	10^{-3}	10^{-2}
0.5	3.8	2.82-5	1.33-4	6.47-4	3.57-3	2.63-2
	4.7	3.02-5	1.42-4	6.89-4	3.73-3	2.66-2
	5.3	3.14-5	1.48-4	7.14-4	3.83-3	2.68-2
	---	(2.53-6)	(2.53-5)	(2.53-4)	(2.53-3)	(2.50-2)
1.0	3.8	3.53-5	1.63-4	7.50-4	3.46-3	2.10-2
	4.7	3.78-5	1.75-4	8.06-4	3.70-3	2.15-2
	5.3	3.94-5	1.82-4	8.40-4	3.84-3	2.17-2
	---	(1.92-6)	(1.92-5)	(1.92-4)	(1.91-3)	(1.90-2)
1.5	3.8	4.02-5	1.85-4	8.23-4	3.42-3	1.81-2
	4.7	4.32-5	1.98-4	8.89-4	3.69-3	1.87-2
	5.3	4.49-5	2.07-4	9.29-4	3.86-3	1.90-2
	---	(1.60-6)	(1.60-5)	(1.60-4)	(1.60-3)	(1.60-2)
2.0	3.8	4.42-5	2.02-4	8.82-4	3.38-3	1.63-2
	4.7	4.74-5	2.17-4	9.55-4	3.69-3	1.69-2
	5.3	4.94-5	2.26-4	9.99-4	3.86-3	1.72-2
	---	(1.39-6)	(1.39-5)	(1.39-4)	(1.39-3)	(1.39-2)
2.5	3.8	4.75-5	2.16-4	9.30-4	3.34-3	1.50-2
	4.7	5.10-5	2.33-4	1.01-3	3.68-3	1.57-2
	5.3	5.31-5	2.42-4	1.06-3	3.88-3	1.59-2
	---	(1.25-6)	(1.25-5)	(1.25-4)	(1.25-3)	(1.25-2)
3.0	3.8	5.04-5	2.29-4	9.72-4	3.31-3	1.40-2
	4.7	5.42-5	2.46-4	1.06-3	3.67-3	1.46-2
	5.3	5.64-5	2.58-4	1.11-3	3.89-3	1.49-2
	---	(1.15-6)	(1.15-5)	(1.15-4)	(1.14-3)	(1.14-2)
3.5	3.8	5.30-5	2.40-4	1.01-3	3.29-3	1.32-2
	4.7	5.70-5	2.58-4	1.10-3	3.66-3	1.38-2
	5.3	5.93-5	2.69-4	1.15-3	3.89-3	1.42-2
	---	(1.06-6)	(1.06-5)	(1.06-4)	(1.06-3)	(1.06-2)
4.0	3.8	5.54-5	2.50-4	1.04-3	3.27-3	1.26-2
	4.7	5.95-5	2.69-4	1.14-3	3.67-3	1.32-2
	5.3	6.20-5	2.81-4	1.20-3	3.91-3	1.36-2
	---	(9.96-7)	(9.96-6)	(9.96-5)	(9.95-4)	(9.92-3)

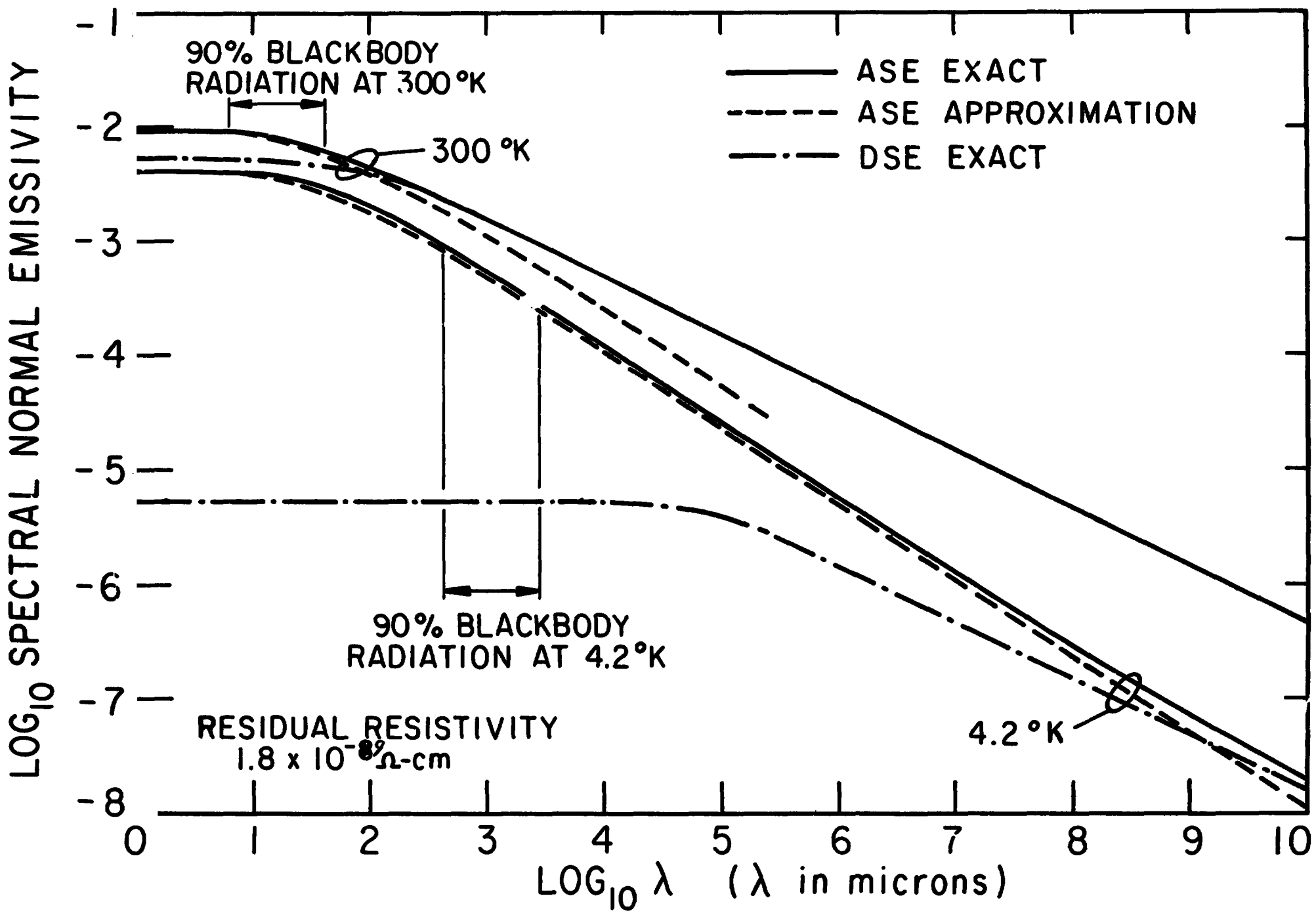

FIG. 1 THEORETICAL SPECTRAL VARIATION OF THE NORMAL EMISSIVITY FOR COPPER

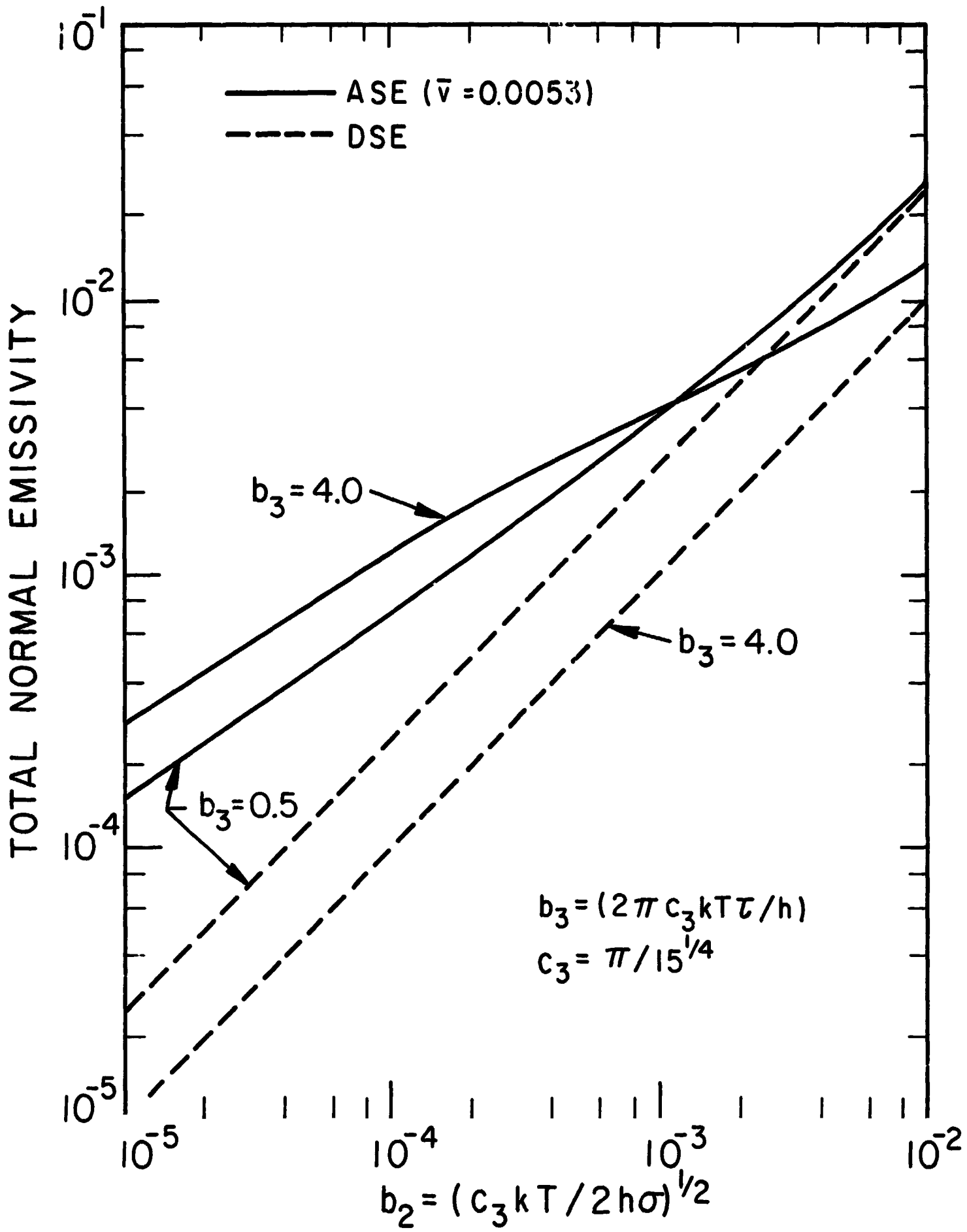

FIG. 2 THEORETICAL TOTAL NORMAL EMISSIVITY OF METALS

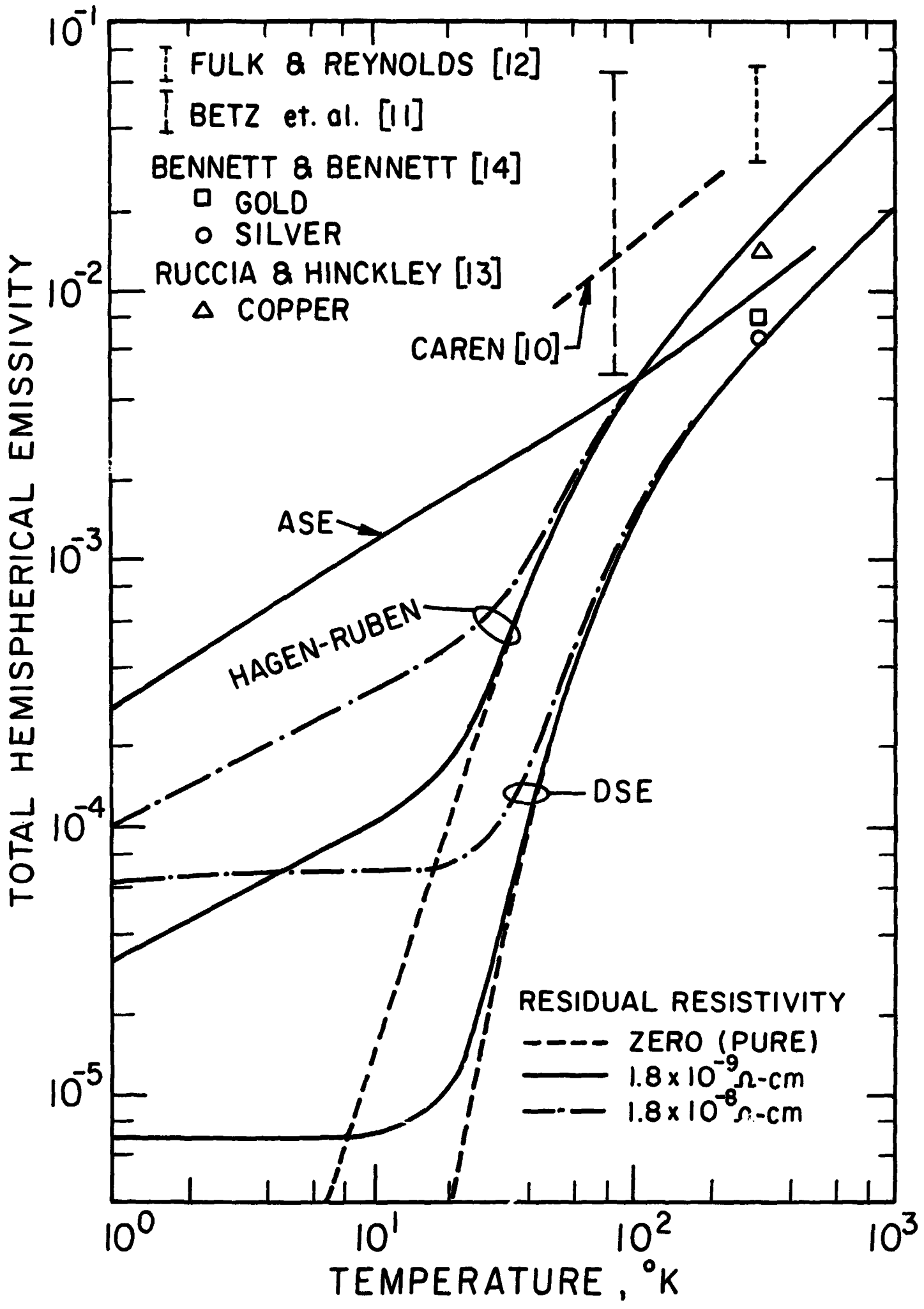

FIG. 3 TOTAL HEMISPHERICAL EMISSIVITY OF COPPER

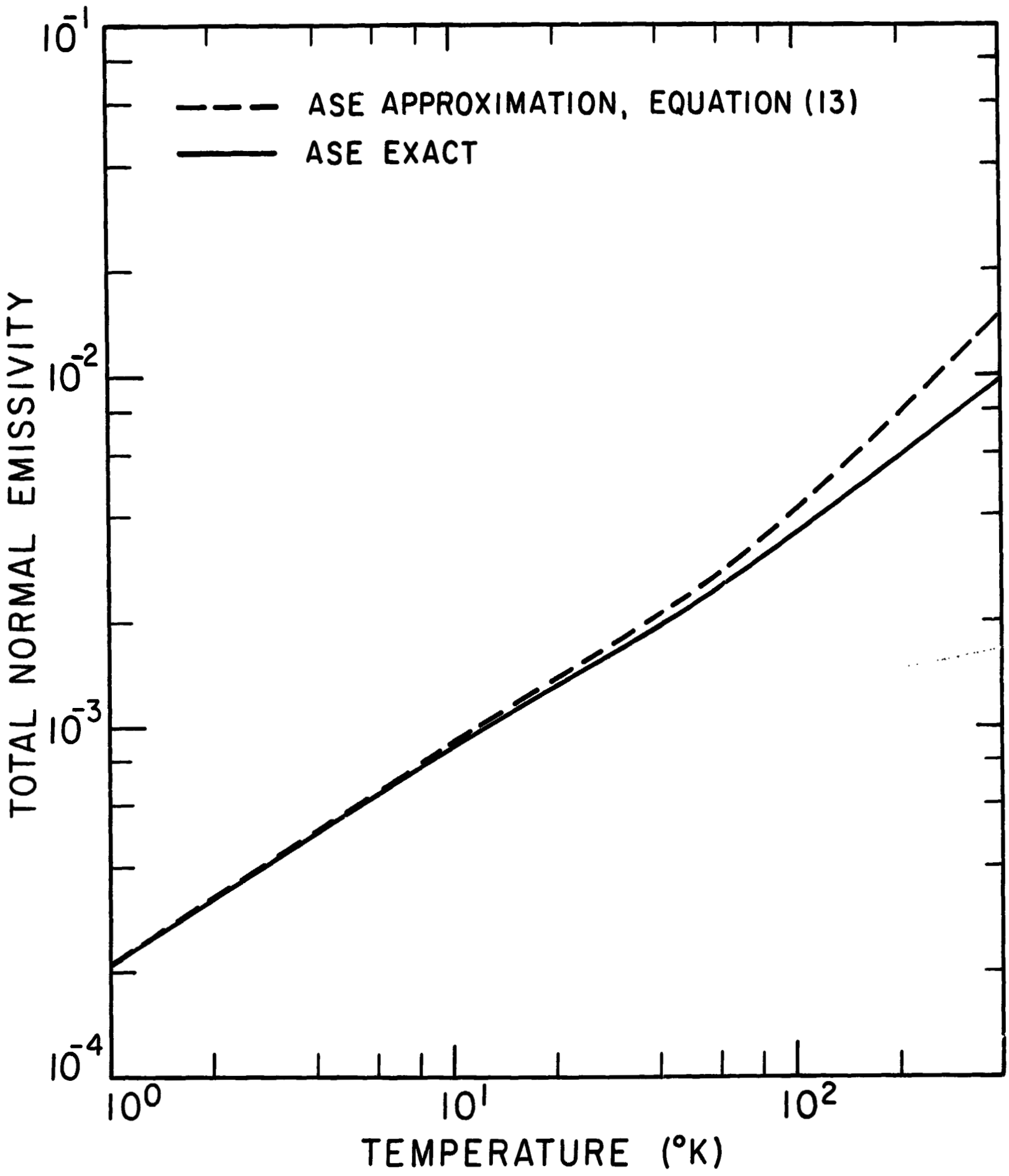

FIG. 4 COMPARISON OF THE ASE APPROXIMATE AND EXACT RESULTS FOR THE TOTAL NORMAL EMISSIVITY OF COPPER