Prediction of Temperature-Dependent Absorptivities of Metallic Materials at 1.06 μm and 10.6 μm

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The optical constants and absorptivity of selected elemental metals and alloys are calculated based on the Drude type model. The absorptivities of the elemental metals at 10.6 μ m agreed with experimental data very well, except for transition metals. Absorptivity values for alloys are calculated by assuming that the sum of contributions of N/m ratio for each constituent metal is equal to the N/m ratio of the alloy. Agreement of alloy and element absorptivity calculated values and experimental data is good at 10.6 μ m but not at 1.06 μ m. Overall the calculation by Drude model gives good estimates of absorptivity at 10.6 μ m.

Nomenclature

= electron electric charge [C] е complex refractive index, dimensionless k m^* optical mass of the electron [kg] mass of the electron [kg] m refractive index, dimensionless n number of free electrons per cubic centimeter Ν = reflectivity, dimensionless R temperature [K] T= permittivity of free space [F m-1] spectral absorptivity, dimensionless α angular frequency of radiation [rad s-1] ω plasma frequency [rad s-1] $\omega_{\rm p}$ relaxation time of electrons [s] τ conductivity of material [ohm-1 m-1] $\sigma_{\rm o}$ damping frequency [s-1] γ = resistivity of material [ohm-m] ρ = wavelength of incident radiation [µm]

I. Introduction

Any model for laser processing of materials must have a complete description of the coupling between the laser source and the material. The coupling is defined by the spectral absorptivity of the material at the wavelength of operation; hence the normal spectral absorptivity is a critical parameter of interest for many applications in this area¹. Optical properties of bulk metals are typically functions of wavelength, temperature, surface geometry (roughness), incident intensity, and physical atomic structural and electrical properties of the material. Since absorptivity is dependent on temperature, it is also important to consider the absorptivity change in the modeling of laser heating².

The wavelength-dependent absorptivity data were found in Ref. 3-5. The temperature-dependent absorptivity data are very few⁶, possibly due to the difficulty in conducting temperature dependent experiments. The

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experimental study by Wieting and Schriempf⁷ provides the only temperature-dependence absorptivity data available for alloys. Zhang and Modest⁸ presented experimental results on temperature-dependent absorptances of ceramics for Nd:YAG and CO2 laser processing applications. Temperature-dependent absorptivity of many metallic materials at the temperature near its melting point is not directly available in the existing literature.

Although obtaining the absorptivity value from experimental investigation is preferred, calculation of the absorptivity based on a theoretical model is also important for the situation when the absorptivity is not readily available for a particular material of interest. One method to predict the absorptivity of an electromagnetic field that obeys Maxwell's equations, is to use the following equation based on the Fresnel reflection relation

$$\alpha = 1 - R = 1 - \frac{(n-1)^2 + k^2}{(n+1)^2 + k^2}$$
 (1)

where n and k are the real and imaginary parts of refractive index, which can be determined using various models.

The Drude theory is a modified oscillator type model developed for reflection and absorption estimation and has been used by many researches⁹⁻¹². This model uses the electrical properties of the material and optical properties of the material. The Hagen-Ruben relation is a model, which provides ease of calculation and can be applied for frequencies much less than the mean collision rate of the electrons in the metal¹³. Sokolov¹⁴ presented a method of determining optical properties of alloys, with a modified damping function. Dausinger and Shen⁶ provided temperature dependent models based on the Drude model. Weaver et al. ¹⁵ presented a compilation of data sets for a number of practical metals.

The temperature-dependent absorptivity of selected pure metals and alloys at infrared regions of the electromagnetic spectrum were calculated and will be presented and compared with the existing experimental data. The absorptivity values of the selected metals are of necessity for accurate laser processing models.

II. Predictive Methods

1. Drude Theory

Prediction of absorptivity using eq. (1) requires knowledge of the real and imaginary parts of the refractive index. One widely used model is Drude theory, which predicts the absorptivity of conducting materials in the infrared and visible wavelengths using a free electron model⁹⁻¹². The equations of the Drude theory are expressed as

$$n^{2} - k^{2} = 1 - \omega_{n}^{2}(\omega + \tau^{-2})$$
 (2)

$$2nk = \omega_p^2 / [\omega \tau (\omega^2 + \tau^{-2})] \tag{3}$$

When both n and k are real and positive, Eqs. (2-3) can be solved to yield¹⁰.

$$n = \frac{1}{\sqrt{2}} \left\{ \left[(1 - Q)^2 + \left(\frac{Q}{\omega \tau} \right)^2 \right]^{1/2} - Q + 1 \right\}^{1/2}$$
 (4)

$$k = \frac{1}{\sqrt{2}} \left\{ \left[(1 - Q)^2 + \left(\frac{Q}{\omega \tau} \right)^2 \right]^{1/2} + Q - 1 \right\}^{1/2}$$
 (5)

Where

$$Q = \frac{\omega^2_p}{\omega^2 + \tau^{-2}} \tag{6}$$

The plasma frequency can be expressed as Ref. 10

$$\omega_p^2 = \left(\frac{Ne^2}{m^* \varepsilon_0}\right)^{1/2} \tag{7}$$

and the relaxation time, or electron lifetime is

$$\tau = \left(\frac{m * \sigma_0}{Ne^2}\right) \tag{8}$$

Damping frequency is inversely related to relaxation time, i.e.,

$$\gamma = \frac{1}{\tau} \tag{9}$$

2. Alloys

The absorptivities for alloys will also be calculated using Drude theory. It is assumed that each constituent metal is contributing the total free electrons according to its concentration in the alloy, so that parameters N/m^* and γ for the alloys can be approximated by summing the contributions of each group, even though these alloys have complex Fermi surfaces and substantial structure disorder⁷. In addition, the absorptivities of alloy are not sensitive to N/m^* , and the ratio can be approximated by assuming $m^*=m$ in Ref. 7. For the case when an optical electron mass was available this was used in the calculation, otherwise the free electron mass was used, as in the case with Aluminum $(m^*=1.32 \times 10-30 \text{ kg})$.

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III. Absorptivities of Pure Metals

Ordal et al. 9 collected optical constants data from the literature, and calculated the Drude parameters ω_p and γ for six metals to see if the experimental data fit to the Drude model. Their results show that the transition metals did not fit with the Drude model except tungsten, and that noble metals fit well. Arnold 10 obtained absorptivities of Ag, Au, Al, Cu, Pb, and W at 10.6 μ m using optical data in Ref. 9 and Drude theory. The predicted absorptivities of Ag, Au, Al, and Cu at room temperature were compared with the available experimental data but no comparison was made at elevated temperature.

Based on Arnold's method¹⁰, the Drude parameters and optical constants were calculated using Eqs. (2)-(3), then the temperature-dependent absorptivities were computed for three metals Al, Cu, and Ni, using temperature-dependent conductivity data and the results are summarized in Tables 1-3. The conductivity values in Tables 1-3 that were used in calculating Drude parameters, were calculated from the resistivity values obtained from Ref. 17. These calculated absorptivity values were plotted with experimental data⁴ in Figs. 1-3. The lines of the least square fit, in Figs. 1-2 were given by Ref.10. Due to the scarcity of temperature-dependent absorptivity data in literature, only a few experimental data were plotted for comparison (Figs. 1-3). Also as a note, as seen in Figures 1-3 there is inconsistency among the experimental data. This is probably because the experimental data for the pure metals were from different researchers and placed together, which would naturally produce some inconsistency among the results.

Table 1. Calculated Absorptivity at 10.6µm for Aluminum

Temperature (K)	Conductivity σ $x10^{7}$ (mho m ⁻¹)	Resistivity ρ x 10 ⁻⁸ (ohm m)	Plasma freq. $\omega_p x 10^{16}$ (rad/sec)	Relaxation time $\tau x 10^{-14}$ (sec)	n	k	Reflectivity	Absorptivity
200	2.660	2.722	1.002	1.040	25.70	101.66	0.001	0.000
300	3.660	2.733	1.992	1.040	25.70	101.66	0.991	0.009
400	2.580	3.87	1.992	0.734	31.94	94.38	0.987	0.013
500	2.000	4.99	1.992	0.569	35.78	87.29	0.984	0.016
600	1.630	6.13	1.992	0.464	38.01	80.70	0.981	0.019
700	1.360	7.35	1.992	0.387	39.16	74.53	0.978	0.022
800	1.150	8.70	1.992	0.327	39.52	68.73	0.975	0.025
900	0.982	10.18	1.992	0.279	39.29	63.41	0.972	0.028

Table 2. Calculated Absorptivityat 10.6 μm of Copper.

Temperature (K)	Conductivity σ $x10^{7}$ (mho m ⁻¹)	Resistivity ρ x 10 ⁻⁸ (ohm m)	Plasma freq. $\omega_p x 10^{16}$ (rad/sec)	Relaxation time tx10 ⁻¹⁴ (sec)	n	k	Reflectivity	Absorptivity
300	5.80	1.73	1.6355	2.45	10.10	89.62	1.00	0.005
400	4.16	2.40	1.6355	1.76	13.66	88.11	0.99	0.007
500	3.24	3.09	1.6355	1.37	16.94	86.23	0.99	0.009
600	2.64	3.79	1.6355	1.11	19.90	84.03	0.99	0.011
700	2.22	4.51	1.6355	0.94	22.51	81.59	0.99	0.012
800	1.90	5.26	1.6355	0.80	24.77	78.95	0.99	0.014
900	1.66	6.04	1.6355	0.70	26.70	76.19	0.98	0.016

Table 3. Calculated Absorptivity at $10.6 \mu m$ for Nickel.

Temperature (K)	Conductivity σ $x10^{7}$ (mho m ⁻¹)	Resistivity ρ x 10 ⁻⁸ (ohm m)	Plasma freq. $\omega_p x 10^{16}$ (rad/sec)	Relaxation time τx10 ⁻¹⁴ (sec)	n	k	Reflectivity	Absorptivity
300	1.39	7.2	0.3221	15.12	0.008	2.444	0.996	0.004
400	0.847	11.8	0.3221	9.223	0.008	2.444	0.990	0.004
500	0.565	17.7	0.3221	6.149	0.019	2.444	0.989	0.007

600	0.392	25.5	0.3221	4.268	0.027	2.444	0.984	0.016
700	0.312	32.1	0.3221	3.39	0.034	2.443	0.980	0.020
800	0.282	35.5	0.3221	3.066	0.038	2.443	0.978	0.022
900	0.259	38.6	0.3221	2.819	0.041	2.443	0.976	0.024

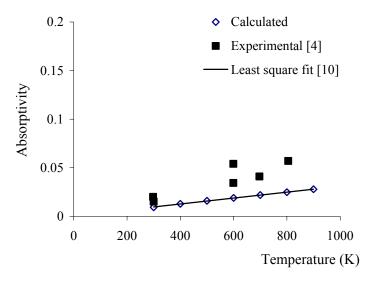


Figure 1. Calculated and experimental temperature-dependent absorptivity of aluminum (10.6 μm).

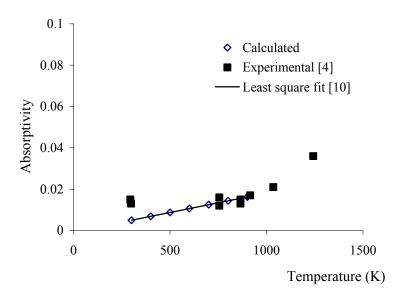


Figure 2. Calculated Temperature-dependent Absorptivity of Copper using Drude parameters (10.6 μm).

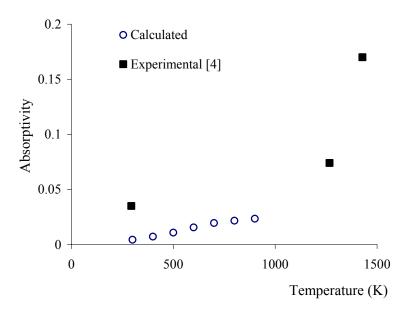


Figure 3. Calculated and Experimental Absorptivity for Nickel (10.6 µm)

It is seen from Figs. 1-3 that the calculated absorptivities are lower than the experimental values. Since surface roughness significantly influences the absorptivity values⁶⁻⁷, this could account for the higher absorptivity values in the experimental data. Considerable deviation is observed in the Nickel data as shown in Fig. 3. Transition metals are known for the deviation from the Drude model, because the interband absorption takes place even at lower energy⁶. Noble metals, on the other hand, are known to fit the Drude model better than the transition metals⁶.

Therefore, the deviation in Nickel absorptivity values from the calculated values could be caused by the interband contribution.

IV. Absorptivities of Alloys

We calculated the absorptivity of AISI 304 stainless steel and Ti-6Al-4V over a range of wavelengths, and tabulated and plotted the results in Tables 4-5 and Figs. 4-5. Also, plotted in Fig. 4 are the experimental results from Ref. 4 and Ref. 7. The calculated absorptivity of the major component in AISI 304 – Fe is also shown in Fig. 4. The absorptivity of AISI 304 is higher than that of pure iron metal, which is in agreement with Dausinger and Shen⁶. The temperature-dependent absorptivity of AISI 304 at 10.6 µm is also calculated and the results are given in Table 5 and Fig. 5. Theoretical values of AISI 304 were calculated based on the resistivity for High alloy steel En 58A302S25 (composition: 0.08% C, 0.3-0.5%Mn, 8% Ni, 18-20% Cr, balance Fe), because temperature-dependent resistivity data for AISI 304 (0.08% C, 19% Cr, 2% Mn, 10% Ni, 1% Si) were not available. The experimental results from Ref. 7 are also plotted in Fig. 5 for comparison. It can be seen from Figs. 4 and 5 that our calculated results agreed very well with the experimental data. It also can be seen that the absorptivity of alloys increases slowly with increase in temperature, which was the direct result of the conductivity in alloys varying diminutively with temperature.

Table 4. Calculation of Wavelength-dependent Drude Parameters and Optical Constants for the AISI 304

N/m* x 10 ⁵⁹ (kg ⁻¹ m ⁻¹)	Resistivity (ohm m) $\Omega \times 10^{-7}$	Conductivity (ohm m ⁻¹) $\sigma x 10^6$	Plas ma freq. $(Ne^2/m^*\epsilon_0)^{1/2}$ $\omega_p x 10^{16}$	Relaxation time $(m^*\sigma_0/Ne^2)$ $\tau x 10^{-16}$	Wavelength (μm) λ	n	k	Reflectivity	Absorptivity
1.77084	7.20	1.39	2.27	3.06	0.1	0.184	0.662	0.599	0.401
1.77084	7.20	1.39	2.27	3.06	0.5	1.863	4.804	0.762	0.238
1.77084	7.20	1.39	2.27	3.06	1.00	4.281	7.303	0.789	0.211
1.77084	7.20	1.39	2.27	3.06	2.00	7.632	10.075	0.827	0.173
1.77084	7.20	1.39	2.27	3.06	3.00	9.997	12.049	0.850	0.150
1.77084	7.20	1.39	2.27	3.06	4.00	11.907	13.702	0.866	0.134
1.77084	7.20	1.39	2.27	3.06	5.00	13.549	15.162	0.877	0.123
1.77084	7.20	1.39	2.27	3.06	6.00	15.012	16.488	0.886	0.114
1.77084	7.20	1.39	2.27	3.06	7.00	16.343	17.712	0.894	0.106
1.77084	7.20	1.39	2.27	3.06	8.00	17.573	18.855	0.900	0.100
1.77084	7.20	1.39	2.27	3.06	9.00	18.722	19.931	0.905	0.095
1.77084	7.20	1.39	2.27	3.06	10.00	19.804	20.952	0.909	0.091
1.77084	7.20	1.39	2.27	3.06	11.00	20.830	21.925	0.913	0.087
1.77084	7.20	1.39	2.27	3.06	12.00	21.807	22.856	0.916	0.084

Table 5. Calculation of Temperature-dependent Drude Parameters and Optical Constants for the AISI 304 at 10.6 µm.

Temp. (K)	$N/m^* \times 10^{59}$ (kg ⁻¹ m ⁻¹)	Resistivity (ohm m) $\Omega \times 10^{-7}$	Conductivity (ohm m ⁻¹) $\sigma x 10^6$	Plasma freq. $ (Ne^2/m^*\epsilon_0)^{1/2} $ $ \omega_p \times 10^{16} $	Relaxation time $(m*\sigma_0/Ne^2)$ $\tau \times 10^{-16}$	n	k	Reflectivity	Absorptivity
298	1.77084	7.20	1.39	2.27	3.06	20.349	21.468	0.911	0.089
373	1.77084	7.76	1.29	2.27	2.84	19.645	20.642	0.908	0.092
473	1.77084	8.50	1.18	2.27	2.59	18.817	19.682	0.904	0.096
673	1.77084	9.76	1.02	2.27	2.25	17.619	18.316	0.897	0.103
873	1.77084	10.72	0.93	2.27	2.05	16.846	17.446	0.892	0.108
1073	1.77084	11.41	0.88	2.27	1.93	16.349	16.892	0.888	0.112
1273	1.77084	11.96	0.84	2.27	1.84	15.983	16.486	0.886	0.114

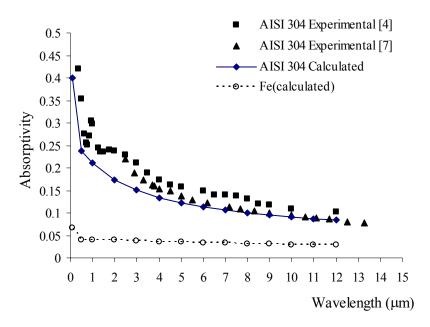


Figure 4. Wavelength-dependent Absorptivity for AISI 304 Stainless Steel.

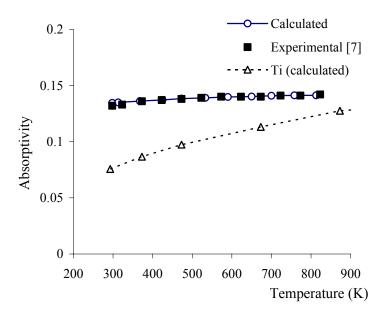


Figure 5. Temperature-dependent Absorptivity for Titanium Alloy Ti-6Al-4V (10.6 μm).

The method of calculating the absorptivity for Tables 4-7 uses temperature-dependent resistivity data of the alloy (Ref. 3) and elemental properties of the constituents. With Eq. (7), the plasma frequency of the metal can be determined. For the alloys as shown in Tables (4)- (7), the summation of the N/m^* ratio is used as the N/m^* value for the alloy. Temperature-dependent resistivity data were used to calculate the damping frequency using Eq. (8) as a function of temperature. These values were used in a straightforward manner for calculation of the complex

refractive index, using Eqs. (4)-(6), from which the reflectivity can be determined with the Fresnel reflection relation. For Inconel (Table 6), the mass of an electron was used as m^* for the calculation except for Ni, where the proper effective mass was used. Also, the calculated results are based on temperature-dependent resistivity data for Inconel with a small Carbon amount (composition: 79.5 Ni, 13.0 Cr, 6.5 Fe, and 0.08 C). In Table 7, the electron mass was used to estimate m^* values, except for Al, Cu, Ni, and Zn. For Al 7075, the small component (i.e. Cr=0.3%) was omitted in the calculation.

Table 6. Calculated Temperature-dependent Drude Parameters and Optical Constants of Inconel at 10.6 μm and 1.06 μm

Elements	Elemental N/m* x 10 ⁵⁹ Note (1) (kg ⁻¹ m ⁻³)	Fractional Contribution N/m* x 10 ⁵⁹ (kg ⁻¹ m ⁻³)	Resistivity (ohm m) Ω x 10 ⁻⁷	Conductivity (mho m ⁻¹) $\sigma \times 10^6$	Relaxation time $(m*\sigma_0/Ne^2)$ $\tau x 10^{-16}$	n	k	Reflectivity	Absorptivity
Ni	1.0026	0.7971							
Cr	1.8282	0.2377							
Fe	1.8642	0.1212							
C	2.1465	0.0017							
Estimated va	alues at 10.6 μm	1							
	Temp(K)								
	390.0	1.1577	9.85	1.0152	3.4171	17.34	18.40	0.8972	0.1028
	573.2	1.1577	10.00	1.0000	3.3658	17.22	18.26	0.8965	0.1035
	774.1	1.1577	10.10	0.9901	3.3325	17.14	18.16	0.8959	0.1041
	872.0	1.1577	10.15	0.9852	3.3161	17.10	18.11	0.8957	0.1043
Estimated va	alues at 1.06 μm	ı							
	Temp(K)								
	390.0	1.1577	9.85	1.0152	3.4171	3.53	6.28	0.765	0.235
	573.2	1.1577	10.00	1.0000	3.3658	3.53	6.23	0.762	0.238
	774.1	1.1577	10.10	0.9901	3.3325	3.53	6.20	0.761	0.239
	872.0	1.1577	10.15	0.9852	3.3161	3.53	6.19	0.760	0.240

Note (1) mass of an electron was used for the calculation except for Ni.

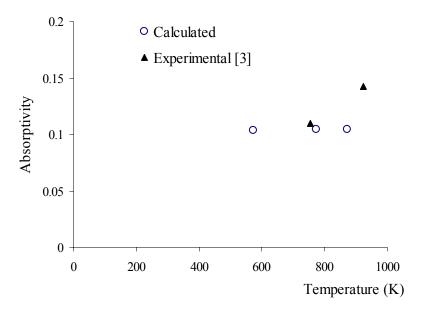


Figure 6. Calculated and Experimental Temperature-Dependent Absorptivity of Inconel at $10.6 \mu m$.

Table 7. Calculation of Drude Parameters and Optical Constants at Room Temperature for Selected Alloys at 10.6 µm.

Elements	Elemental N/m* x10 ⁵⁹ (kg ⁻¹ m ⁻¹)	Fractional Contribution N/m*x10 ⁵⁹ (kg ⁻¹ m ⁻¹)	Resistivity (ohm m) Ω x10 ⁻⁸	Conductivity (mho m ⁻¹) $\sigma \times 10^7$	Plasma freq. $(Ne2/m*\epsilon_0)^{1/2}$ $\omega p \times 10^{16}$	Relaxation time $(m*\sigma_0/Ne^2)$ $\tau x 10^{-16}$	n	k	Reflectivity	Absorptivity
Ti B120 VA	C (Ti-V-Cr-Al)									
Ti	1.2540	0.9154								
V	1.5858	0.2062								
Cr	1.8282	0.2011								
Al	1.3681	0.0410								
Estimated va	lues at 10.6µm	1.3637	149.00	0.0671	1.9882	1.9176	14.3120	14.7768	0.8736	0.1264
Estimated va	lues at 1.06µm	1.3637	149.00	0.0671	1.9882	1.9176	3.6319	5.0102	0.6880	0.3120
A-110-AT (T	(i-Al-Sn)									
Ti	1.2540	1.1599								
Al	1.3681	0.0684								
Sn	0.6404	0.0160								
Estimated va	lues at 10.6µm	1.2444	163.00	0.0613	1.8992	1.9211	13.6847	14.1268	0.8682	0.1318
Estimated va	lues at 1.06µm	1.2444	163.00	0.0613	1.8992	1.9211	3.4750	4.7848	0.6761	0.3239
Al 2024										
Al	1.3681	1.2778								
Cu	0.9229	0.0415								
Mg	0.9454	0.0142								
Mn	0.8814	0.0053								
Estimated va	lues at 10.6µm	1.3388	5.33	1.8762	1.9699	54.6046	35.6974	84.7985	0.9833	0.0167
Estimated va	lues at 1.06µm	1.3388	5.33	1.8762	1.9699	54.6046	0.5262	10.5623	0.9815	0.0185
Al 7075										
Al	1.3681	1.2381								
Zn	1.6968	0.0933								
Mg	0.9454	0.0236								
Cu	0.9229	0.0138								
	lues at 10.6µm	1.3689	5.38	1.8587	1.9920	52.9066	36.4720	84.7617	0.9830	0.0170
Estimated va	lues at 1.06µm	1.3689	5.38	1.8587	1.9920	52.9066	0.5489	10.6790	0.9811	0.0189

The calculated results are also compared with experimental data in Figs. 6-10. Figure 6 shows Inconel absorptivity versus temperature and it is seen that the calculated data have little variation due to the nearly constant resistivity values. Experimental data for Inconel at the wavelengths of interest were difficult to find, however a few temperature dependent points show that a reasonable approximation was found by the Drude model. Figure 7, shows a good estimate for A-110-AT at 10.6 μm, and however at 1.06 μm the variation in experimental values used does not give a consistent picture for prediction at this wavelength for the model. For Fig. 8, at 10.6 μm the prediction by Drude theory is good, and there were no experimental values to compare at 1.06 μm. In Figs. 9 and 10 experimental and calculated match well at 10.6 μm, and though at 1.06 μm a difference of about 0.1 in absorptivity is shown. For Figs. 7-10, the agreement between calculated values and experimental data is good at 10.6 μm, but experimental values are much greater than those expected from the theory at 1.06 μm in all the results. The higher absorption can be contributed by parallel band absorption, as shown by Ashcroft and Sturm¹⁶. This band does not appear in the Drude absorption theory. As shown in the case of aluminum, the parallel-band absorption occurs at a higher energy than the Drude theory predicts.

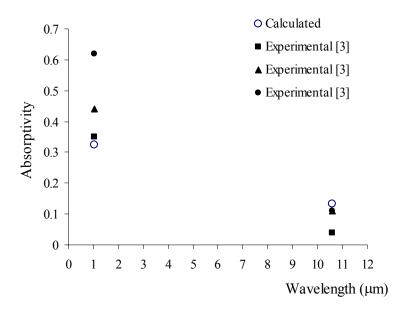


Figure 7. Calculated and Experimental Absorptivity for A-110-AT

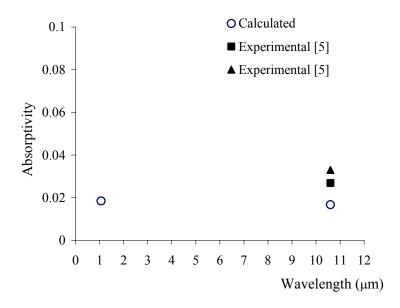


Figure 8. Calculated and Experimental Absorptivity for Al 2024.

V. Conclusion

The absorptivities of several selected pure metals and alloys were predicted using Drude theory. The predicted absorptivities of the pure metals at $10.6~\mu m$ agreed very well with the experimental results except for the transition metals. Agreement of alloy and element absorptivity calculated values and the experimental data is good at $10.6~\mu m$ but not at $1.06~\mu m$. For practical purposes the Drude type model works qualitatively well for alloys at $10.6~\mu m$.

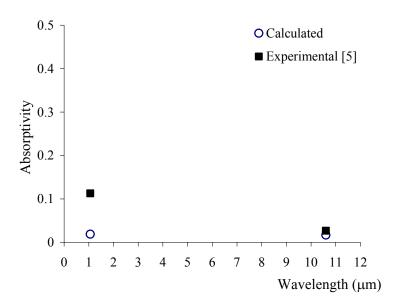


Figure 9. Calculated and Experimental Absorptivity for Al 7075.

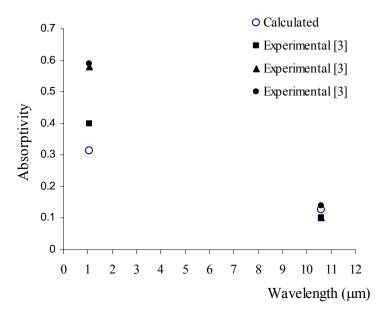


Figure 10. Calculated and Experimental Absorptivity for Ti B120 VAC

Acknowledgments

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