REFLECTIVITY OF NAK DROPLETS

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ABSTRACT

An important contribution to the space debris population near 900 km orbital altitude are the NaK droplets. Sixteen nuclear powered satellites of the type RORSAT launched between 1980 and 1988 activated a reactor core ejection system close to this altitude. The core ejection causes an opening of the primary coolant circuit. The liquid coolant has been released into space during these core ejections, forming droplets up to a diameter of about 5.5 cm. These droplets consist of an alloy of two alkali metals, sodium and potassium (NaK). In this paper the monochromatic and the total reflectivity of NaK is calculated using theoretical models. The reflectivity depends on the alloy composition and temperature of a droplet. The alloy composition may change due to evaporation, resulting in an enrichment of sodium especially at the droplet surface. According to the literature, there is only a limited number of available measurement data concerning the optical properties of NaK alloys. Furthermore the published data for pure sodium and potassium are controversial. Thus it is necessary to investigate the optical properties of alkali metals and their alloys. Mainly two types of optical absorption, the intraband and the interband absorption, are considered. The intraband absorption is calculated using the Drudemodel which uses electrical properties to derive the optical constants of pure metals or alloys. Drude assumes that the valence electrons can be treated as free electrons. The electrons behave like an ideal gas of uncharged particles. The theory of free electrons is a very simple model for the description of the valence electrons in metals. This assumption is sufficient for alkali metals, because they show a nearly free electron behavior. For the interband absorption the classical Butcher-model is used. Furthermore an absorption anomaly which has been observed in some alkali metals is discussed. Especially for potassium, some measurements revealed an unexpected absorption in the visible and the near infrared. This absorption can be determined using a model according to Overhauser who assumes charge-density waves (CDW). But the existence of the anomalous absorption is controversial. The influence of the different models on the reflectivity is discussed. The reflectivity is calculated depending on the alloy composition at the surface of a droplet. The results are compared to measurement data from the literature. It is shown that NaK droplets have a very high total reflectivity.

1. INTRODUCTION

NaK droplets are massive metal objects with a spherical shape and a size up to 5.67 cm diameter. If the surface is purely metallic, the droplets should have a very high reflectivity. Values of the total reflectivity are given in the literature (s. Tab. 1).

Reflectivity R	Reference	
0.84 - 0.89	Sridharan [80]	
0.85 ± 0.07	Hall [31]	

Tab. 1. Measured reflectivity of orbiting NaK droplets.

The alkali metals like sodium and potassium have a very simple atomic structure which makes it easier to describe their physical properties using theoretical approaches. Havstad [35] estimated the optical constants of liquid metals on the basis of the Drude-theory and confirmed the applicability of this theory for liquid metal alloys. But a detailed literature review revealed that further absorption mechanisms must be considered in the visible and near infrared region. Beside the intraband absorption (Drude-absorption), there appears an interband absorption which means a transfer of an electron to a higher band [60, 83].

To calculate the optical properties of orbital NaK droplets, it is necessary to know the alloy composition at the surface. Generally, sodium and potassium have a high solubility in each other and should not show any significant tendency towards segregation [14, 29]. But there exist several theoretical investigations which

indicate that a segregation can occur [6, 7, 9, 13, 42, 44, 45, 46, 70, 71, 72, 74]. In this case, it would be likely that potassium is segregated to the surface due to its lower surface tension, compared to sodium. (The driving force behind surface segregation is the reduction of the surface energy.) But the vapor pressure of potassium is higher than that of sodium, resulting in a higher evaporation rate. This may cause an enrichment of sodium in the surface of a droplet [53, 54, 62]. Consequently it is difficult to predict the exact alloy composition at the surface of a NaK droplet.

2. INTRABAND ABSORPTION

The literature mentions a theory for the calculation of the optical constants of metals which is qualified especially for alkali metals [16, 25, 27, 30, 39, 56, 64, 85]. This theory was developed about the year 1900 by Paul Drude, who derived the optical constants from the electrical properties of metals. Drude assumed that the valence electrons can be treated as free electrons. The band structure and the periodic structure of the crystal potentials were neglected. The electrons behave like an ideal gas of uncharged particles. The model of free electrons is a very simple model for the description of the valence electrons in metals. The Drude-theory is a good approximation for metals with simple structures. The Drude-model is qualified for alkali metals, because their valence electrons show a nearly free behavior. The optical constants can be derived simply from the Hall-coefficient and the specific electrical resistance. The Hall-coefficient is a measure for the electron density, and the resistance correlates with the average time between collisions. Both depend on the alloy composition. Havstad [35] showed that published values of the Hall-coefficient and the electrical resistance of liquid metal alloys can provide useful estimations of the optical constants for some binary alloys.

The optical properties are expressed as complex dielectric constant ε' . The parameters and the used notation is given in the following [19, 33, 37].

$$\varepsilon' = \varepsilon_1 + i\varepsilon_2 \tag{1}$$

$$\varepsilon_1 = 1 - \frac{4\pi e^2 \tau^2}{m_e (1 + \omega^2 \tau^2)} = 1 - \frac{N_e e^2 \tau^2}{m_e \varepsilon_0 (1 + \omega^2 \tau^2)} = n^2 - k^2$$
(2)

$$\varepsilon_2 = \frac{4\pi e^2 \tau}{m_e \omega (1+\omega^2 \tau^2)} = \frac{N_e e^2 \tau}{m_e \varepsilon_0 \omega (1+\omega^2 \tau^2)} = 2nk \tag{3}$$

The following parameters are used: elementary charge e [1.6e10-19 C], extinction coefficient k, mass of an electron m_e [kg], index of refraction n, permittivity of free space ε_0 [8.85e-12 F/m], real part ε_1 and imaginary part of the complex dielectric function ε_2 , and angular frequency ω [1/s]. The Drude-model can be applied by computing the number density N_e [1/m³] and the average time between collisions τ [s] from measurements of the Hall coefficient R_H [m³/(As)] and the direct current resistance ρ [Ω m]. R_H and ρ depend on the alloy composition.

$$N_e = 1/(R_H e)$$
 , $\tau = m_e /(N_e e^2 \rho)$ (4)

The monochromatic (wavelength depending) reflectivity R_{λ} can be calculated from the optical constants. (Sometimes the absorption index κ instead of the extinction coefficient is used.)

$$n = \left[\frac{1}{2}\left(\sqrt{\varepsilon_1^2 + \varepsilon_2^2} + \varepsilon_1\right)\right]^{0.5} \tag{5}$$

$$k = \left[\frac{1}{2}\left(\sqrt{\varepsilon_1^2 + \varepsilon_2^2} - \varepsilon_1\right)\right]^{0.5} = n\kappa$$
(6)

$$R_{\lambda} = \frac{(n-1)^2 + k^2}{(n+1)^2 + k^2} = \frac{n^2(1+\kappa^2) + 1 - 2n}{n^2(1+\kappa^2) + 1 + 2n}$$
(7)

The total reflectivity *R* can be derived by weighting R_{λ} with the solar spectrum or the black body temperature and integration over all wavelengths.

3. INTERBAND ABSORPTION

The interband absorption appears in the visible and the UV region in addition to the Drude-absorption. Interband absorption means the transfer of electrons from the conduction band into energetic higher, unoccupied bands. The imaginary part of the complex dielectric function ε_2 is a measure for the absorption. The overall absorption is calculated by adding the interband absorption $\varepsilon_{2,ib}$ to the Drude-absorption $\varepsilon_{2,D}$.

$$\varepsilon_2 = \varepsilon_{2,D} + \varepsilon_{2,ib} = 2nk \tag{8}$$

The absorption is mostly expressed as optical conductivity σ , where v is the frequency in [1/s].

$$\sigma = nk\nu = \frac{1}{2}\varepsilon_2\nu \qquad , \nu = \frac{\omega}{2\pi} \tag{9}$$

There exists a standard theory of interband absorption by Butcher [10] and Meessen [52]. The optical conductivity of the interband absorption σ_{ib} is calculated with Eq. (10-12) [10, 66, 73, 76].

$$\sigma_{ib} = \frac{m_e e^2}{\pi \hbar^4} \frac{|V_{110}|^2}{G_{110}} \frac{(\omega - \omega_0)(\omega_1 - \omega)}{\omega^3} , \text{ for } \omega_0 < \omega < \omega_1 , \hbar = \frac{h}{2\pi}$$
(10)

$$G_{110} = \frac{2\pi\sqrt{2}}{a} \quad , \ a = \left(\frac{2}{N_e}\right)^{1/3} \tag{11}$$

$$\omega_0 = \frac{\hbar}{2m_e} (G_{110} - 2k_F) G_{110} \qquad , \ \omega_1 = \frac{\hbar}{2m_e} (G_{110} + 2k_F) G_{110} \qquad , \ k_F = (3\pi^2 N_e)^{1/3}$$
(12)

The following parameters are used: Fourier component of the pseudopotential V_{110} [J], reciprocal lattice vector G_{110} [1/m], threshold and cutoff frequencies ω_0 and ω_1 [1/s], Planck's constant *h* [6.6262e-34 Js], lattice constant *a* [m], and Fermi-radius k_F [1/m]. The optical conductivity σ in Eq. (8) uses CGS esu units and is expressed in [1/s], while the optical conductivity of the interband absorption σ_{ib} in Eq. (10) uses SI units [1/(Ω m)]. Both units can be converted using 1/(Ω m) = 1e-11 c^2 /s, where *c* is the speed of light expressed in [cm/s] [61]. But this theoretical model leaves one parameter open which is determined by fitting to experimental results [4]. This is the Fourier component of the pseudopotential V_{110} . The values of V_{110} which were found in the literature are summarized in Tab. 2.

Tab. 2. Summarized values of the Fourier component of the pseudopotential V_{110} , given in the literature.

$ V_{110} $ of sodium	Reference	$ V_{110} $ of potassium	Reference
[eV]		[eV]	
0.11	Ham [32]	0.08	Gibbons [28]
0.18	Ashcroft [5]	0.158	Harms [33]
0.225	Overhauser [66]	0.163	Dresselhaus [15]
0.23	Gibbons [28]	0.195	Overhauser [66]
0.323	Butcher [10]	0.23	Ashcroft [5]
0.44	Phillips [69]	0.24	Ham [32]
0.475	Miskovsky [57]	0.305	Butcher [10]

Values for V_{110} were chosen which show the best fitting of the calculated interband absorption to measured data, especially in the visible region, where the solar spectrum has its maximum intensity. For sodium and potassium values of $V_{110,Na} = 0.32$ eV and $V_{110,K} = 0.23$ eV according to Butcher [10] and Ashcroft [5] were selected. The results are shown in Fig. 1 and 2. There was no theoretical approach for the treatment of alloys found in the literature. Thus the interband absorption was approximated by interpolating V_{110} depending on the alloy composition. The number density N_e was approximated by using the Hall coefficient R_H from Eq. (4).

4. ABSORPTION ANOMALY

The discussion about an unexpected absorption of alkali metals began in 1962 with a work of El Naby [19], who investigated the optical properties of potassium between wavelengths from $\lambda = 0.365 \,\mu\text{m}$ to 2.0 μm . The results were also published in [18, 47, 48] and were later reviewed by Harms [33] with new experiments. The unexpected result of El Naby was the detection of a strong maximum in the absorption at $\lambda = 1,6 \,\mu\text{m}$. A similar behavior was measured by Hietel [37] for sodium in 1965. This unexpected absorption peak was later called "absorption anomaly" [33]. (Harms could not reproduce the absorption peak measured by El Naby.)

The results of these measurements motivated several theoretical and experimental studies. The literature gives only a limited sets of data concerning the optical properties of massive alkali metal samples. Most experiments were made with thin alkali films. The experiments of Hietel and El Naby belong to the very few which were made for bulk materials. To follow the discussing of the results of theses studies and to find more measurement data, a detailed literature review was performed. This bibliographical study comes to the result, the optical constants of the alkali metals are still controversial. Most work was done in the fields of the monochromatic optical properties of pure sodium or potassium. In the literature several theoretical and experimental results are given for sodium [2, 4, 11, 21, 22, 26, 34, 36, 37, 41, 53, 66, 75, 82]. Other studies give results for potassium [1, 3, 8, 10, 17, 20, 24, 38, 49, 52, 58, 68, 73, 77, 78, 81, 85, 86, 88]. Only one reference was found, where NaK-alloys were investigated [59]. Unfortunately these results refer to only one selected wavelength.

In the literature it is discussed whether the measured optical properties are dominated by volume or surface effects [50, 55, 78, 84]. The optical properties vary with the film thickness [8, 40, 51, 63, 82]. Thus measurements which have been made with bulk materials are more suitable for the estimation of the optical properties of NaK droplets. There are fundamental differences between films and bulk materials. Films may have a reduced electrical conductivity [79]. Surface effects may occur for rough surfaces [82]. This means, thin films consisting of solid metal may have grain boundaries or lattice defects which influence the surface properties [40]. Differences in the surface roughness may produce different experimental results [8]. The surface of liquid metals is smooth [82]. Therefore it is not expected that surface effects have an influence on the absorption behavior of liquid metals. If the NaK droplets are liquid at the surface and no impurities are segregated to the surface, then the optical properties are defined by volume effects and should comply with the properties of bulk materials.

The absorption anomaly measured by El Naby [19] is very high. The additional absorption occurs below the interband threshold. This anomalous peak cannot be explained by the classical theory [88]. The peak appears at wavelengths where the theoretical absorption should be low. The additional absorption was observed in the solid as well as in the liquid state. Such a behavior was only detected for the alkali metals [12]. In the literature it is discussed, whether the classical model of interband absorption may be too simple [66]. Overhauser [65, 67] postulated a new interband transfer mechanism, caused by spin-density waves (SDW) or charge-density waves (CDW). The experimental data of El Naby are in remarkable good agreement with this theory. Although the CDW-theory provides a good approximation of the absorption anomaly, the existence of these wave has not been verified [33].

Other experiments do not show anomalous effects and confirm the classical theory. Generally there should be simply intra- and interband absorption and no anomalous peaks. Experiments by Smith [76, 77, 78], made with sodium, showed no effects which disagree with the theory. According to Bennett [8] the experimental situation was cleared by the measurements of Smith, and the traditional view was reestablished. However, also these measurement results are not unambiguous. Indeed, Smith's results revealed no unexpected peaks, but the Drude-absorption is higher than predicted by the theory [88]. The absorption is only qualitatively but not quantitatively in good agreement with the theory. Smith worked with films of sodium. In contrast, the measurements for sodium and potassium by Hietel [37] and El Naby [19] were made with bulk materials. But also the measurement results for potassium are doubtful. Harms [33] reviewed the results of El Naby and could not reproduce the anomalous peak. According to Harms, the measured optical constants of El Naby must be attributed to a contaminated or eventually rough potassium surface.

5. OPTICAL CONSTANTS

The most important published measurement data of sodium and potassium are shown in Fig. 1 and 2. The data are compared with the Drude- and interband theories. The data of potassium are also compared to the CDW-theory given by Overhauser. Both diagrams show the absorption expressed as optical conductivity σ depending on the frequency (photon energy hv) using Eq. (9). The optical conductivity is typically in use in such diagrams all-over the physical literature, because it points out the interband absorption in a pronounced manner. The boundaries of the visible region are shown. Therein the wavelength of $\lambda = 546.1$ nm is presented, where Morgan

made his measurements with NaK, varying the alloy composition. For the calculation of the optical conductivity, electrical quantities from [23] were used.



Optical Conductivity of Sodium

Fig. 1. Optical conductivity of sodium at room temperature. Comparison of the Drude- and interband (IB) models with measured data from Hietel [37], Palmer [68], Duncan [17], Smith [78] and Monin [58]. The wavelength $\lambda = 546.1$ nm is pointed out, where Morgan [59] gives data for varying alloy compositions.



Optical Conductivity of Potassium

Fig. 1. Optical conductivity of potassium at room temperature. Comparison of the Drude- and interband (IB) as well as CDW-models with measured data from El Naby [19], Palmer [68], Duncan [17], Smith [78], Harms [33] and Whang [86]. The wavelength $\lambda = 546.1$ nm is pointed out, where Morgan [59] gives data for varying alloy compositions.

Fig. 1 and 2 show that in the visible region many experimental data deviate from the Drude-model. Most experts attribute this to the interband absorption. In the infrared region the Drude-model is an appropriate approximation. In Fig. 2 the remarkable absorption anomaly measured by El Naby is compared to the CDW-model according to

Overhauser. Whether the absorption anomaly is attributed to charge-density waves or to surface contamination is discussed controversially in the literature. Due to the fact that most experimental studies did not confirm the existence of an anomalous peak, the CDW-model will not be used in the following.

Pure sodium or potassium are solid at room temperature, while the eutectic alloy NaK-78 (77.8 weight % of potassium) remains liquid. In the liquid state an evaporation of an orbiting droplet is possible [87]. The alloy component potassium would evaporate faster, due to its high vapor pressure compared to sodium. So it may be possible that the alloy composition at the surface of a droplet is enriched with sodium. If the surface solidifies, for example when all potassium has evaporated from the outer layers of a droplet, the evaporation process may be interrupted. If the surface composition of a droplet can vary, then it is necessary to estimate the optical properties of NaK depending on the alloy composition. The calculation of the optical constants is based on the following simplifying assumptions. The Hall-coefficient R_H and the direct current resistance ρ are calculated depending on the alloy composition. Furthermore V_{110} is linearly interpolated between the values of pure sodium and potassium. Some material values are also determined by extrapolation or interpolation, for example in two phase regions. These approximations result in a relatively smooth variation of the optical constants with the alloy composition. This result is justified by investigations of Kent [43] who found that the optical constants of solid or liquid metallic alloys with varying alloy composition can be approximated with smooth, non-linear curves. The optical constants of NaK are calculated and compared to measurement data of Morgan [59]. These data were exclusively measured at one selected wavelength of $\lambda = 546.1$ nm and are the only data for NaK alloys which were found in the literature. The result is shown in Fig. 3.



Fig. 3. Imaginary part of the complex dielectric function ε_2 as measure for the absorption behavior of NaK at room temperature depending on the alloy composition. Comparison of the model for intraband (Drude) plus interband absorption with measurement data from Morgan [59] at a wavelength of $\lambda = 546.1$ nm. The vertical line marks the eutectic point.

In Fig. 3 the measurement data of Morgan [59] are compared to the sum of intraband and interband absorption. for NaK alloys. The comparison shows that the curve of the model is qualitatively in good agreement with the data. Quantitatively the model predicts a lower absorption as given by the data. A possible explanation may be an imperfectness of the surface [18] used by Morgan.

The total reflectivity R can is calculated by weighting R_{λ} from Eq. (7) with the solar spectrum and integration over all wavelengths. The results is shown in Fig. 4.



Fig. 4. Total reflectivity *R* of of NaK at room temperature depending on the alloy composition, calculated with the model for intraband (Drude) plus interband absorption. The vertical line marks the eutectic point.

Fig. 4. Shows that the reflectivity R varies between 92 % and 98 % depending on the alloy composition. This more than the value of roughly 85 % given by Sridharan and Hall for orbital NaK droplets (s. Tab. 1).

6. CONCLUSION

In this work the optical constants of sodium and potassium are calculated using theoretical models for the intraband and interband absorption given by Drude and Butcher. The optical constants of NaK are approximated on the basis of these models depending on the alloy composition. The results are in qualitatively good agreement with measurement data. But most data show a quantitatively deviation from the theory, especially in the visible region. If this deviation is explained by surface contamination or roughness, then the model gives the maximum possible reflectivity for a clean and smooth surface. It may be possible that the effective reflectivity of orbital NaK droplets is lower than the calculated values of 92 % to 98 % due to surface contamination, segregation of impurities to the surface, or surface roughness. The CDW-theory given by Overhauser is not considered, because of the controversial views in the literature concerning the existence of charge-density waves. Furthermore the this theory is only for potassium in good agreement with the measurement data, not for sodium.

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