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LOW ENERGY INTERBAND TRANSITIONS IN ALUMINIUM

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Résumé. — Les spectres de thermoréfectance de monocristaux d'aluminium présentent des structures à 0,5 et 1,5 eV. L'analyse de ces spectres donne la variation avec la température des coefficients $V(111)$ et $V(200)$ du pseudopotential. Le modèle à deux bandes parallèles est inadéquat pour expliquer complètement les spectres observés. Une étude de la variation avec l'énergie des coefficients du pseudopotential semble possible.

Abstract. — Thermorefectance spectra of single crystals of aluminium show very clearly transitions at 0.5 and 1.5 eV. Analysis of the spectra yields the temperature variation of the pseudo-potential coefficients V_{111} and V_{200} . Discrepancies between the data and the two parallel band model point to possible studies of the energy variation of these coefficients.

Aluminium is certainly the polyvalent metal whose optical properties have been most thoroughly studied [1]. The spectrum is rather flat, dominated by a strong absorption peak near 1.5 eV and a much weaker one around 0.5 eV. This last structure was observed only quite recently with a calorimetric technique by Bos and Lynch [2]. These two structures are qualitatively well described theoretically. In polyvalent metals, there are parallel bands where the Fermi surface cuts the Brillouin zone. Partly empty, partly occupied throughout a region of k space, such parallel bands lead to sharp peaks in the conductivity at a photon energy of $2|V_G|$, where V_G is the appropriate Fourier coefficient of the pseudo-potential [3-5]. Here we report on new thermorefectance measurements which show clearly the two structures and give the variation between helium and room temperature of the pseudo-potential coefficients. Our experiments complement earlier data by Rosei and Lynch [6], they had observed the $2|V_{200}|$ transition at 1.5 eV but failed to detect the $2|V_{111}|$ transition.

The samples were prepared from sheets 2 mm thick of 99.98 % aluminium. The sheet was then unidirectionally rolled to 0.25 mm. The temperature of the

metal was then raised very slowly to 515 °C and it was annealed at that temperature for 72 hours to secure a good recrystallization [7]. The aluminium was very slowly brought back to room temperature, and the samples were then electropolished to the desired thickness of about 0.1 mm. The samples were in the form of large single crystals with perhaps 10^7 cm^{-3} defects. The number of defects is drastically raised when the sample is clamped at the cold tip of a variable temperature cryostat. The surface of the sample is formed by a 10 Å layer of amorphous Al_2O_3 , and complexes of Cl and P [7].

The temperature of the sample is modulated by electron bombardement [8], we can feed up to one watt of modulating power. In this technique, the sample is not strained against a substrate and this partly explains why we observed a large signal at 0.5 eV, besides the fact we have good sized mono-crystals.

Our data is seen in figure 1. With a good signal to noise ratio, the two peaks are clearly seen and we have followed their temperature evolution from about helium temperature to room temperature. As in ref. [6], there is no sign of any structure at 0.8, 1.2 and 1.45 as predicted and reported [9], neither do we see the fine structure on the 1.5 eV transition recently reported [10]. Perhaps could we only mention the

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FIG. 1. — Thermoreflectance spectra of a 100 μ single crystal sample of aluminium, at 10 K and 300 K. The low energy data reads on the 5×10^{-5} scale, the high energy ones on the 4×10^{-4} scale. On the 10 K data is shown a typical fit with the parallel band model.

extra hump between 1.1 and 1.3 eV at room temperature.

In these data, as opposed to Rosei and Lynch who also measured the thermotransmission, we have measured only the thermoreflectance and we have difficulties in quantitatively interpreting the data. It could be quite misleading to make a Kramers Krönig analysis of the $\Delta R/R$ spectrum since we have not measured the optical constants of our samples. The best thing to do was to see how far we could go with the parallel bands analysis. At any given temperature, the interband conductivity from transitions between parallel bands is given by Ashcroft and Strum [5] and depends on $|V_G|$ and τ_G a phenomenological interband relaxation time. With the Drude conductivity characterized by a relaxation time τ_0 and the plasma frequency ω_p , we have therefore six parameters, each in principle temperature dependent. We have calculated the reflection coefficient from two sets of coefficients, hopefully at two slightly different temperatures and computed $\Delta R/R$. The starting values were taken from published data [9], in particular from Mathewson and Myers [11], the variations of the parameters were adjusted to optimize the fit. In no case was the agreement very good over the full experimental range. ΔV_G is felt on the low energy side of the transition while $\Delta \tau_G$ gives comparable amplitudes on both side of the transition but the

rapid return to zero about 1.7 eV cannot be obtained. The two structures can be adjusted more or less independently. The non zero averaged value of $\Delta R/R$ is given by $\Delta \tau_0/\tau_0$.

Over a large domain of reasonable values for the parameters, whatever the chosen values for τ_G and $\Delta \tau_G$, the inflexion point between the maximum and the minimum occurs quite accurately at $2|V_G|$ when the parallel bands model is used and thus, we have an accurate determination of these two parameters as given in table I for 4, 77 and 300 K, the accuracy being much greater for $|V_{200}|$ than for $|V_{111}|$. This inaccuracy is reflected in the apparent insensitivity of V_{111} at low temperatures. The pseudo-potential coefficients have a marked variation with temperature, they are products of i) a form factor which depends on temperature through the thermal expansion and the dielectric function and ii) a structure factor with a Debye-Waller temperature dependence. Mathewson and Myers [11] have analysed their data in these terms, and we give in table I the zero temperature values deduced from such a reduction of the data.

TABLE I

	V_{111} (eV)	V_{200} (eV)	
4 K	0.225 ± 0.005	0.753 ± 0.002	present data
	0.243	0.764	dHvA, ref. [12]
	0.988	0.985	correction factor, ref. [11]
0 K	0.228 ± 0.005	0.764 ± 0.002	present corrected
77 K	0.225 ± 0.003	0.745 ± 0.002	present
120 K	0.216	0.772	ref. [6]
300 K	0.185 ± 0.01	0.715 ± 0.002	present
	0.190	0.738	ref. [11]
	0.22	0.76	ref. [4]
	0.944	0.932	correction factor, ref. [11]
0 K	0.196 ± 0.01	0.767 ± 0.002	present corrected

In the table, we also give other determinations of the V_{111} and V_{200} . Mathewson and Myers [11] and Motulevitch [4] determine the parameters from fits to the conductivity data. Rosei and Lynch [6] with thermoderivative data have determinations from $\Delta \epsilon_2$, but have also the difficulties mentioned here due to inaccurate absolute values of the optical constants, and having not actually observed the structure at 0.5 eV, they, and all the other workers, deduced the values of V_{111} from data a little outside the sensitive range.

A significant comparison is made with the values of the parameters deduced by Ashcroft [12] in an analysis of the low temperature dHvA data. The agreement is very good for V_{200} but our value for V_{111} is quite smaller and perhaps it is a good indication of the inadequacy of our analysis.

There are two ways of improving the theoretical expression to compare to the data :

1) to consider more than two bands in the pseudo-potential calculation. The difference as shown by

Ashcroft are small and lead to the fine structure of Hunderi and Nilsson, which is not observed here;

2) departures from parallelism and energy dependent pseudo-potential coefficients. Sturm and Ashcroft [13] have studied these points and analysing the data from Mathewson and Myers [11] concluded that an analysis of the optical data could yield this energy dependence. The simple two band model used here fails chiefly in that it leads to too wide structures but our lack of absolute accuracy prevents further analysis. Perhaps the good agreement between the different values of V_{200} is misleading and the main result is the

definite discrepancy between the values of V_{111} determined from analysis of the dHvA data and the optical data. If the energy dependence of V_{111} is significant, the two values should differ as emphasized in ref. [13].

Our data show that optical measurements can not only provide estimates of pseudo-potential coefficients, but high precision data could yield information on their energy dependence provided absolute measurements of the optical constants are made in conjunction with differential measurements.

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