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# Reflection Spectrum of Solid Argon in the Vacuum Ultraviolet

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# REFLECTION SPECTRUM OF SOLID ARGON IN THE VACUUM ULTRAVIOLET[≠]

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*The reflectance of solid Ar has been measured at $20^{o}$ K for an angle of incidence of $15^{o}$ in the photon energy range from 10 to 30 eV using the synchrotron radiation of DESY. The reflectance data reveal a spin orbit split exciton series with sharp maxima converging to about 14 eV together with broader peaks above 14 eV due to transitions between valence and conduction band. The results are compared to the absorption spectrum associated with the 2p core levels.*

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Several optical[1,2] and electron energy loss[3] measurements have been performed on solid Ar in order to study its electronic transitions from the valence band. These investigations were confined to the spectral region below 14 eV. They were made with limited resolution so that the existence of an exciton series converging to the band gap could not clearly be proved. Thus these measurements led to some uncertainty in the determination of the series limit[4].

We have measured the reflectance of solid Ar at 20° K for an angle of incidence of 15° in the energy range 10 to 30 eV with a resolution of 2 Å. The synchrotron radiation of the 7.5 GeV electron synchrotron DESY[5] was monochromatized by a normal incidence monochromator in a modified Wadsworth mounting[6]. The Ar was evaporated as a thin film of unknown thickness onto a KCl single crystal and a glass plate, cooled down in a He-cryostat. No significant influence of the two substrates on the reflectance could be observed. The light reflected from the Ar surface was detected with an open photomultiplier Bendix M 306. Cryostat and multiplier were mounted into a modified commercial ultra high vacuum system. Outside the cooled cryostat the chamber pressure was $5 \times 10^{-8}$ Torr.

Figures 1 and 2 show the results of our measurements. Fig. 1 shows in an extended scale the fine structure up to 14 eV together with results obtained earlier by Baldini[2] and by Bostanjoglo and Schmidt[3]. The reflectance is given in arbitrary units. The internal consistency of the relative height of the different peaks is better than 5 %. While the spectral distribution of the light emerging from the exit slit of the monochromator was taken into account no absolute calibration was made during the measurements. A rough estimation based on the KCl reflectivity at room temperature[7] gave about 70 % for the reflectivity of the highest peak at 12.5 eV, but this value may be incorrect by a factor of 2.

At 12.10 eV we see in Fig. 1 the first peak in very good agreement with the other authors[2,3]. The second peak is relatively broad. The pronounced shoulder at 12.35 eV coincides with Baldini's peak at 12.3 eV[2], the maximum at 12.50 eV with the energy loss maximum of Bostanjoglo and Schmidt[3] at 12.46 eV. Other sharp peaks up to 14 eV can clearly be resolved.

Figure 2 shows broader peaks occuring at higher energies. At 27.5 eV a minimum in the reflectivity curve can be seen, which agrees in its energy position with a "window" line, found in the absorption of solid Ar[8].

We ascribe the sharp peaks in the reflectance below 14.5 eV to two exciton series, namely two Rydberg-series from the spin-orbit split valence band to the bottom of the conduction band at $\Gamma_1$[9,10]. Our assignment for the different peaks to members of the two Rydberg-series, according to the equation[11] $E = E_o - \frac{G}{n^2}$ , is given in Table I, where E is the peak energy, $E_o$ the series limit, G the binding energy and n the quantum number. As the first exciton may not necessarily fit into the Rydberg-formula[11] $E_0$ and G have been evaluated from the n=2 and n=3 members. Reevaluation of the n=1 exciton from these values gives an energy 0.25 eV below the experimental values. This on the other hand explains the disagreement of our G and $E_0$ values with those of other authors[2-4] as they were only able to calculate their values from the n=1 and n=2 excitons.

The two n=1 excitons show a temperature dependence: The peak at 12.35 - 12.5 eV shows slightly varying ratios of the contribution of the two parts and the peak at 12.1 eV splits into two. A detailed investigation of these effects is in progress.

The structures above 14.5 (Fig. 2 upper part) indicate the onset of interband transitions, but no simple correlation with singularities in the conduction band structure obtained by Mattheis[10] seems possible.

The lower part of Fig. 2 shows for comparison the absorption coefficient due to 2p transitions in solid Ar around 250 eV.[12] There is an obvious similarity between 3p and 2p transitions. Both spectra begin with a sharp line followed by a broad asymmetric one and by several weaker peaks. If one attributes the first lines of the 2p absorption spectrum to an exciton series, too, one is led to assume much larger binding energies for 2p excitons than for 3p excitons. The high energy parts of the spectra, consisting presumably of broad maxima due to transitions into the conduction band continuum, reflect further similarity. A conversion of the reflectance data into $\varepsilon_2$ by a Kramers-Kronig-analysis should allow a more precise discussion.

We want to thank the directors of our Institutes for their continuing support.

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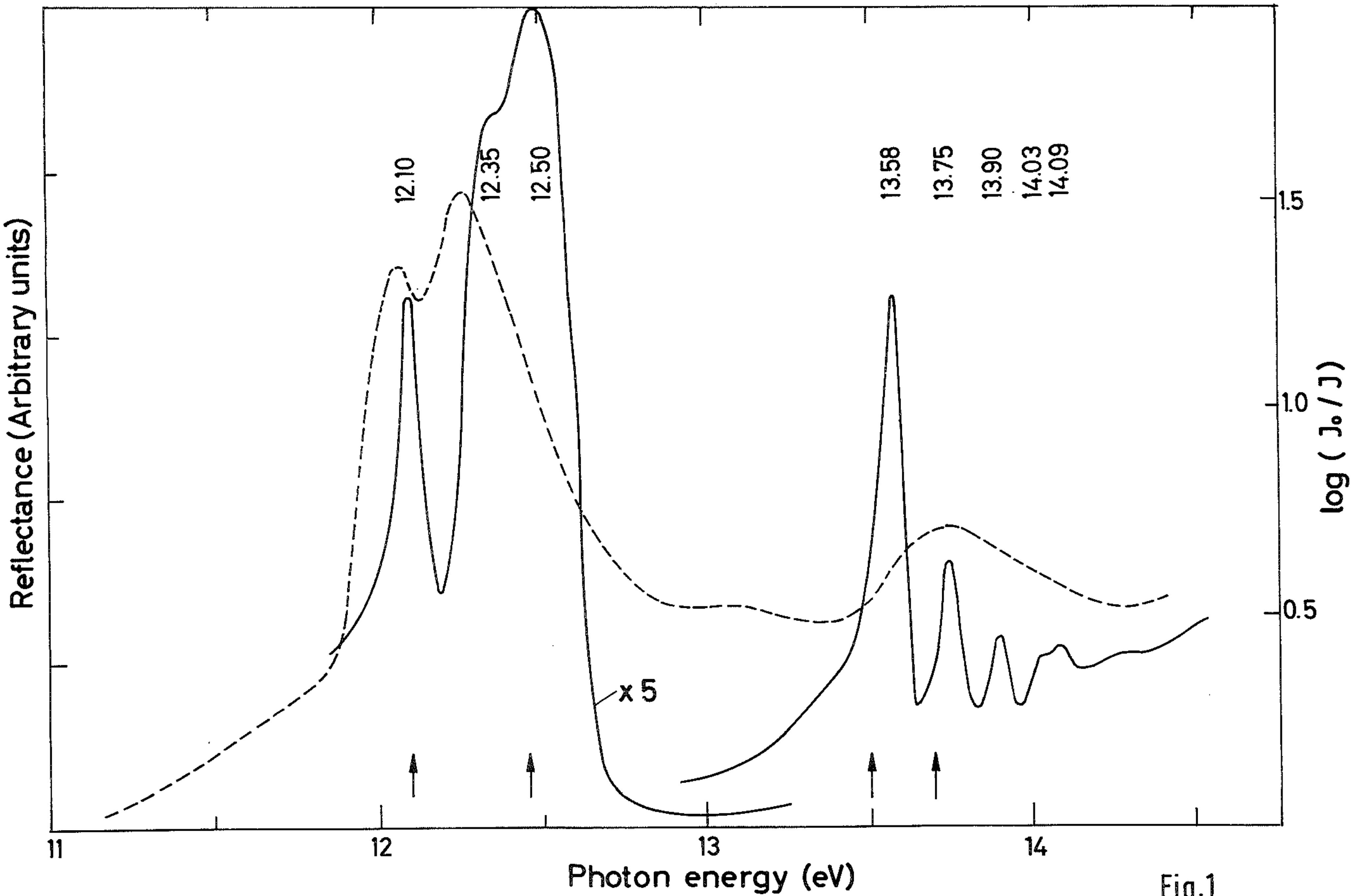
Table I: Energy position and identification of the exciton peaks

| Peak energy eV | Γ (3/2) series | Γ (1/2) series |
|---|---|---|
| 12.10 | n=1 | |
| 12.35 | | n=1 |
| 12.50 | | |
| 13.58 | n=2 | |
| 13.75 | | n=2 |
| 13.90 | n=3 | |
| 14.03 | | n=3 |
| 14.09 | n=4 | |
| G | 2.30 eV | 2.06 eV |
| $E_o$ | 14.16 eV | 14.25 eV |

## Figure captions

1) Reflectance of solid Ar at $20^{\circ}$ K for an angle of incidence of $15^{\circ}$ between 11 and 14.5 eV (solid line). The dashed line gives Baldini's absorption curve[2], right ordinate. Arrows indicate the energy position of electron energy loss peaks obtained by Bostanjoglo and Schmidt[3].

2) Reflectance of solid Ar at $20^{\circ}$ K for an angle of incidence of $15^{\circ}$ between 11 and 30 eV (solid line). Absorption coefficient of solid Ar at $15^{\circ}$ K around 250 eV (dashed line).



Reflectance (Arbitrary units)
log ( J₀ / J )
Photon energy (eV)
12.10
12.35
12.50
13.58
13.75
13.90
14.03
14.09
x 5
11
12
13
14
1.5
1.0
0.5


Fig.1

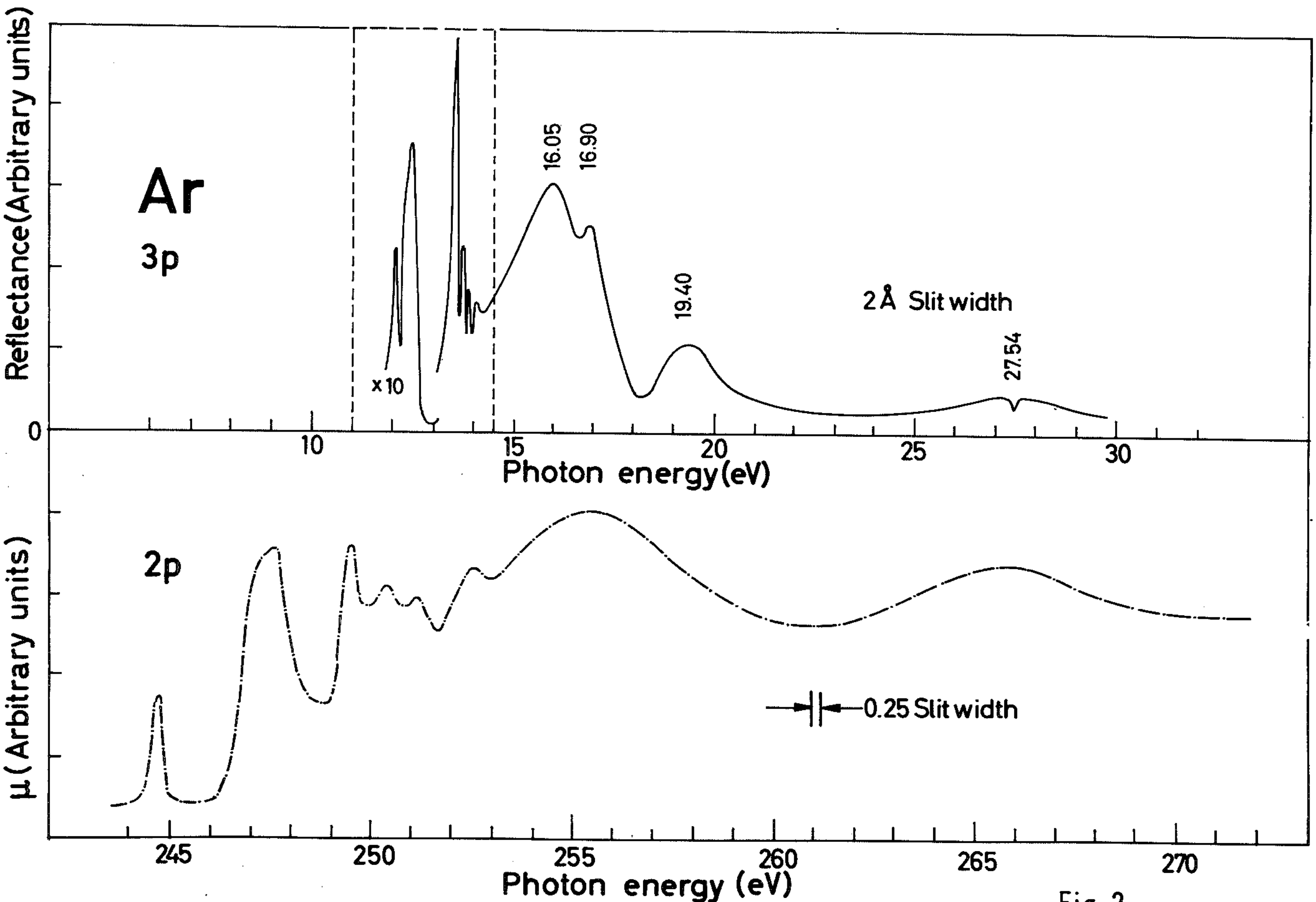

Ar
3p
Reflectance(Arbitrary units)
0
x10
16.05
16.90
19.40
27.54
2 Å Slit width
10
15
20
25
30
Photon energy(eV)
2p
μ (Arbitrary units)
0.25 Slit width
245
250
255
260
265
270
Photon energy (eV)

Fig. 2