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Experimental Comparison of Photoabsorption of
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The photoabsorption structure of thin films of solid xenon has been investigated near the $N_{IV,V}$ -edge (~ 66 eV) and compared with the structure of gaseous Xe. As Baldini found in the fundamental absorption region, exciton-like peaks as well as interband transitions have been found in the solid. The data are discussed in view of recent band calculations.

In the last few years several experimental and theoretical investigations of the optical properties of gaseous and solid xenon have been reported. The most extensive measurements on solid Xe were performed by Baldini,¹ which covered the energy range from the onset of fundamental absorption (~ 8 eV) to 14 eV. These data were subsequently discussed theoretically.^{2,3}

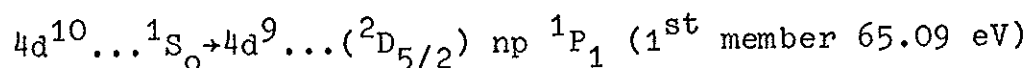
Recently electron energy loss measurements⁴ on solid Xe films were performed up to energies of 22 eV. At higher energies optical measurements have only been performed on Xe gas. Pertinent to the present work are those of Codling and Madden⁵ in the vicinity of the $N_{IV,V}$ -edge of Xe (~ 68 eV). The present note gives our results on optical absorption structure of both solid and gaseous Xe in the energy range 64 - 76 eV.

The results have been obtained by using the synchrotron radiation from the 7.5 GeV electron synchrotron DESY as a radiation source with a continuous spectral distribution.⁶ The spectrograph⁷ was used at a grazing angle of incidence of 300 mrad; this yielded, together with the spectral distribution of the synchrotron radiation, a short wavelength cutoff at about 120 eV, resulting in a negligible intensity of second order light for the wavelength region under investigation. The solid Xe has been prepared in thin films on a zapon substrate in a cryostat. The temperature could be varied down to that of liquid He. By careful shielding, the contamination rate was very low. Intensity loss due to contamination was only a few percent within ten hours.

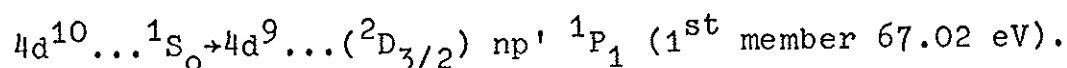
Fig. 1 shows the energy dependence of the absorption coefficient evaluated from measurements on samples of different thickness, which were slowly evaporated at a temperature a few degrees below the condensation point (about 50° K). In other samples, which were evaporated onto substrates at liquid

helium temperature, the structure was less pronounced and peaks F and K were missing. By annealing at a temperature close to the sublimation point, most of the structure was recovered, but the peaks were not as prominent as shown in Fig. 1. As the sample thickness has not been determined, the absorption coefficient is given in arbitrary units with an overall relative accuracy of about 15 %. In adjacent regions of the spectrum the relative accuracy is much better. Those structures, which were clearly reproduced in several measurements are labeled with capital letters. The figure also includes the absorption structure of gaseous Xe. The energy calibration of the spectrometer is based on the values for the energy positions of these gas lines given by Codling and Madden.⁵

Coming from the fundamental absorption region, a steady decrease of absorption was observed, until at about 62,5 eV, where a slow rise of the continuous absorption began, followed by the first faint peak A. Both features are relatively weak, and therefore can be seen only in very thick layers. In the gas spectrum one recognizes two series of lines, caused by the spin-orbit splitting of the 4d shell (1.97 eV, see Ref. 5); namely



and



Similar spin-orbit pairs show up in the solid, namely B-G (energy splitting = 1.96 ± 0.04 eV), C-H (1.80 ± 0.11 eV), D-I (2.00 ± 0.09 eV), F-K (1.89 ± 0.06 eV), and M-N (2.02 ± 0.06 eV). Peak L does not show unambiguously a corresponding spin-orbit partner, but this may be hidden in either I or M.

The two peaks B and G are very close to the first member of the gas absorption series and are of similar width. In analogy with the situation in the fundamental absorption region¹, this gives rise to the assumption that the peaks B and G belong to core excitons with a hole in the 4d shell. The structures C, D, E, F, and H, I, J, K show no obvious relationship with the gas lines. The peaks L, M, and N are in an energy region where the gas has only continuous absorption. This makes evident, that they are of pure solid-state origin. As in gaseous Xe, the absorption rises to a broad maximum near 100 eV (not shown in Fig. 1), which is attributed to delayed d→f transitions. In analogy with the situation in the gas^{8,9} we therefore conclude that the d→f transitions near the threshold are suppressed and the observed structure is due primarily to d→p transitions. In addition, normalization of the solid and gas absorption curves to each other below the onset of 4d transitions (62 eV), which appears to be a quite reasonable procedure, yields integrated oscillator strengths in the energy region 64-76 eV for gas and solid which are equal, within the experimental errors. This would suggest that - though the structure has been completely changed while going from gas to solid - the oscillator strength is retained in that region.

An examination of the gas absorption spectrum in the region of peak A was not possible because the zapon windows of the gas absorption cell did not stand pressures which would have been equivalent to our thickest solid Xe films. Data taken at the highest possible pressures gave no indication for such structure in gaseous Xe.

If one tries to understand the absorption structures of solid Xe in the light of Reilly's band calculations,³ one cannot relate the obviously strong exciton B (or G) to the lowest critical points in the conductionband, because all of them have the wrong symmetry.¹⁰ If they are then ascribed to higher points in the zone, the observation of the weak absorption below peak B - namely peak A - would be intelligible as due to transitions to the lowest conductionbands. This structure is weakened because of the selection rules.

Cooling the films down to liquid helium temperature did not cause any difference in the absorption structure. Especially, the widths of B and G did not decrease. This is not surprising because the width of the gas absorption lines is - as mentioned earlier - of the same order of magnitude, and this width is explained by broadening of the initial state 4d shell due to inner Auger transitions.¹¹

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Figure captions:

Fig. 1 Absorption coefficient versus photon energy for solid (solid curve) and gaseous (dashed curve) xenon in the energy range 64 - 76 eV.

