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Far-infrared lattice absorption in gallium phosphide

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Résumé. — L’absorption en infrarouge lointain dans le réseau de phosphure de gallium a été mesurée en utilisant un spectromètre à transformée de Fourier à 300 et 100 K. Des combinaisons à 2 phonons (soustraction et addition) ont été observées dans la bande spectrale 30-300 cm⁻¹. Les caractéristiques observées sont attribuées, sur la base de paires de phonons proches des limites de la zone de Brillouin, aux points X, L, W et K.

Abstract. — The far-infrared lattice absorption in gallium phosphide was measured using a Fourier-transform spectrometer at 300 and 100 K. Two-phonon combination (difference) and overtone bands were identified in the spectral range 30-300 cm⁻¹. The observed features were assigned on the basis of zone-edge phonon pairs at X, L, W and K points.

1. Introduction.

In the recent years considerable experimental work has been done to obtain zone edge critical point phonons in GaP while using inelastic neutron scattering [1, 2], second order Raman scattering [3-5], and infrared absorption [6-10]. Most of the infrared absorption measurements published so far [7-10] were restricted to the spectral range above about 400 cm⁻¹, i.e. above the Reststrahlen band, where two-phonon summation and overtone bands appear. As far as the present authors know, only one work dealt with the infrared lattice absorption below the Reststrahlen band, in the far-infrared range [6]. In the work of Koteles and Datars [6] the lowest frequency feature recorded in the two-phonon far-infrared absorption spectrum of GaP at 20 K was found at 180 cm⁻¹. In this work most of the prominent features of the absorption spectrum were assigned to pairs of phonons on the hexagonal face of the Brillouin zone while phonon pairs at X and L generally were thought to contribute only minor features in the spectrum. The two-phonon features in the spectral range below the Reststrahlen band have mainly been investigated by second order Raman scattering experiments [3-5].

In the present paper we report the results of far-infrared lattice absorption spectra measurements on GaP. The observed spectral features were interpreted invoking difference combinations and overtones of zone-edge phonons.

2. Experimental procedure.

Far-infrared spectra were measured on thin GaP single crystals with average thickness of 200-400 μm at room temperature and at about 100 K. The spectra were taken in the 10-130 cm⁻¹ and 200-310 cm⁻¹ range with a Grubb-Parsons IS-3 interferometer using the Fourier transform technique. Above about 310 cm⁻¹ our samples were totally absorbing, due to the tail of the Reststrahlen absorption. Between 130 and 200 cm⁻¹ the spectra were too noisy to be meaningful. Spectral resolution was about 5 cm⁻¹ and wave number reproducibility was ± 0.5 cm⁻¹. The spectrometer was calibrated using the lines of water vapour. The samples were supported on polyethylene strip.

3. Results and discussion.

The far-infrared absorption spectrum of GaP at room temperature in the spectral range of 20-130 cm⁻¹ and 200-310 cm⁻¹ is presented in figure 1. The spectrum contains various features (peaks and shoulders) which...
Table 1. — Assignment of the features observed in the two-phonon far-infrared absorption spectrum of gallium phosphide.

<table>
<thead>
<tr>
<th>Feature No</th>
<th>300 K cm⁻¹</th>
<th>100 K cm⁻¹</th>
<th>Assignment</th>
<th>Shape</th>
<th>IR (cm⁻¹)</th>
<th>Raman (cm⁻¹)</th>
<th>Neutron (cm⁻¹)</th>
</tr>
</thead>
<tbody>
<tr>
<td>1</td>
<td>31</td>
<td>37</td>
<td>A₁ - A₁</td>
<td>Peak</td>
<td>P₂ P₃</td>
<td>30</td>
<td>30 ± 5</td>
</tr>
<tr>
<td>2</td>
<td>38</td>
<td></td>
<td>A₁ - TA</td>
<td>Peak</td>
<td>P₃ P₄</td>
<td>43 ?</td>
<td>50 ± 9</td>
</tr>
<tr>
<td>3</td>
<td>62</td>
<td></td>
<td>A₂ - A₂</td>
<td>Peak</td>
<td>P₅ P₆ P₇</td>
<td>62 ?</td>
<td>60 ± 17</td>
</tr>
<tr>
<td>4</td>
<td>74 (*)</td>
<td></td>
<td>TO - LA</td>
<td>Peak</td>
<td>P₀</td>
<td>104</td>
<td>104 ± 9</td>
</tr>
<tr>
<td>5</td>
<td>106</td>
<td>105</td>
<td>X</td>
<td>Peak</td>
<td>P₁</td>
<td>213</td>
<td>210 ± 4</td>
</tr>
<tr>
<td>6</td>
<td>112</td>
<td>113</td>
<td>X - K</td>
<td>Peak</td>
<td>P₅ P₆</td>
<td>234</td>
<td>237 ± 17</td>
</tr>
<tr>
<td>7</td>
<td>219</td>
<td>228</td>
<td>2 TA</td>
<td>P₁</td>
<td></td>
<td>247 ± 5</td>
<td></td>
</tr>
<tr>
<td>8</td>
<td>238</td>
<td></td>
<td>O₁ - A₁</td>
<td>Peak</td>
<td>P₅ P₁</td>
<td>258</td>
<td>257</td>
</tr>
<tr>
<td>10</td>
<td>249</td>
<td></td>
<td>2 A₁</td>
<td>Peak</td>
<td>P₂</td>
<td>288</td>
<td>288 ± 8</td>
</tr>
<tr>
<td>11</td>
<td>261</td>
<td></td>
<td>LO - TA</td>
<td>Peak</td>
<td>P₂</td>
<td>294</td>
<td></td>
</tr>
<tr>
<td>12</td>
<td>292</td>
<td></td>
<td>LO - TA</td>
<td>Peak</td>
<td>P₂</td>
<td>294</td>
<td></td>
</tr>
</tbody>
</table>

(*) Reference 6.
(?) Reference 3, the lines at 43 and 62 cm⁻¹ are marked there with ?
(?) Reference 1 for the phonons at X and L.
(?) Reference 2 for the phonons at W and K.
(?) Probably absorption band of polyethylene.
(?) At W approximately $O_{1} = O_{\text{in}}$

From the twelve features listed in table I four and three lines respectively were assigned to difference or overtone bands originating from the critical points W and X respectively, and one feature to difference band at point L and other one to point K. One feature is thought to originate from the absorption of polyethylene supporting strip. The remaining two features were left unaccounted for.

At 100 K temperature feature No 1 disappeared. At the same temperature features Nos 2, 5 and 6 were seen at 37, 105 and 113 cm⁻¹ respectively. Feature No 4 was present only as a shoulder.

To predict the shapes of relevant features, the experimental phonon dispersion curves obtained in inelastic neutron scattering [1, 2] were examined. Where it was possible the probable shape of the singularity in the two-phonon density of states was determined according to the sign of the second derivative of the experimental $\omega(k)$ curves [11]. Use was also made of the theoretical results of Birman [12] concerning the one-phonon critical points in the related diamond structure crystals.

Features Nos 7, 9, 11 and 12 were also observed by Koteles and Datars [6] in their spectra covering the spectral range down to about 180 cm⁻¹. We think that feature No 7 observed here is an unresolved composition of the otherwise well documented line at around 210-212 cm⁻¹ in the Raman spectra [3, 4], usually ascribed to 2 TA(X-K) in the literature [4-6, 13, 14], and an other peak at around 220 cm⁻¹, which we ascribe to optic and acoustic difference combinations at K and W. Feature No 9 is assigned in
to an unspecified location on the hexagonal face of the Brillouin zone. Feature No 11 is generally assigned to a difference band \( \text{TO}(X \text{ or } L) - \text{TA}(X \text{ or } L) \) [3-6, 13, 14]. The feature at 292 cm\(^{-1}\) was assigned by Weinstein and Piermarini [13, 14], on the basis of pressure dependence of Raman intensities either to 2 TA(K) or 2 \( \text{A}_2 \)(W).

As far as the present authors know all the features below 120 cm\(^{-1}\) are reported here for the first time for absorption. Feature No 1 coincides with a major peak observed in second order Raman scattering experiments [3-5], assigned either to \( \text{LO}(X) - \text{TO}(X) \) [3, 4] or \( \text{LO}(L) - \text{TO}(L) \) [5]. Features Nos 2 and 3 probably correspond to two minor but unidentified peaks seen by Hoff and Irwin [3] in second order Raman spectra. The identification of features Nos 1 and 3 as difference bands arising from critical point W appears to be convincing in the light of the recently published inelastic neutron scattering dispersion curves [2].

Finally it should be mentioned that infrared absorption measurements performed on the same crystals in the spectral range above 400 cm\(^{-1}\) by one of the present authors [17] are also consistently interpreted using among others the phonon energies for critical point W deduced here.

4. Acknowledgments.

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References