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LETTER TO THE EDITOR

The localised vibrational mode two-phonon absorption of carbon in gallium arsenide

L Z Zhang[†], Y C Du[†], B R Zhang[†], Y H Wang[‡], B C Ma[‡] and G G Gin^{†§}

[†] Department of Physics, Peking University, Beijing, People's Republic of China

[‡] Beijing General Research Institute of Non-Ferrous Metals, Beijing, People's Republic of China

[§] CCAST (World Laboratory), PO Box 8730, Beijing, People's Republic of China

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Abstract. It is found that the 1115 and 1157 cm^{-1} absorption bands in GaAs doped intentionally with ^{13}C (measured at 300 K) are manifestations of the localised vibrational mode two-phonon absorption of $^{12}\text{C}_{\text{As}}$ and $^{13}\text{C}_{\text{As}}$. The ratios of the absorption intensity for single-phonon processes to that for two-phonon processes for ^{12}C and ^{13}C in GaAs are about 87 at 300 K and 75 at 10 K, respectively.

The introduction of light elements into semiconductors gives rise to local vibrational modes (LVM) and the corresponding absorption can be observed in the mid-infrared (IR) absorption spectrum [1, 2]. LVM absorption has become an important experimental method; it can be used to identify impurities, to obtain their concentrations and to determine their positions or site symmetries in the lattice. Although a large amount of research has been carried out on LVM absorption, as far as we know no unambiguous LVM two-phonon absorption has been reported to date. A weak band at 1013 cm^{-1} in the infrared absorption spectrum of silicon was observed and attributed to an overtone of the 515 cm^{-1} band related to interstitial oxygen [3]. In fact, the overtone of the 515 cm^{-1} mode should be located at about 1030 cm^{-1} and the 17 cm^{-1} difference is too large to merit this conclusion.

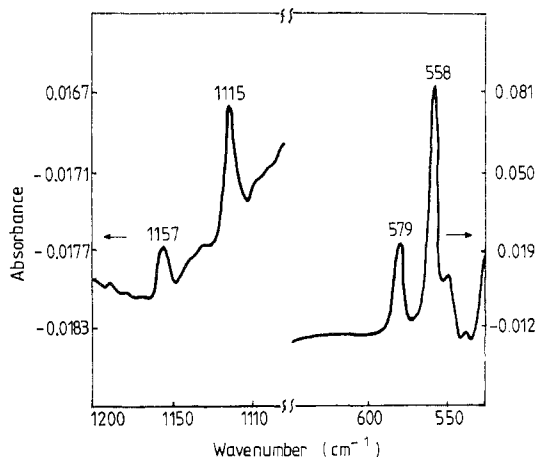
Studying the LVM absorption bands of the prevailing shallow acceptor, carbon, in GaAs, besides the 579 cm^{-1} LVM absorption band attributed commonly to $^{12}\text{C}_{\text{As}}$ in GaAs [4], we have observed a weak 1157 cm^{-1} LVM absorption band with a frequency exactly double that of the former. In order to find out whether the band observed is really the overtone of the LVM absorption band of $^{12}\text{C}_{\text{As}}$ and not an unknown absorption band coincidentally located there, semi-insulating GaAs was intentionally doped with ^{13}C and the existence of LVM two-phonon absorption bands related to both ^{12}C and ^{13}C was confirmed from the absorption measurements.

Three kinds of GaAs sample grown by the liquid-encapsulated Czochralski (LEC) method in a pyrolytic boron nitride (PBN) crucible were used. The parameters of all the samples used are listed in table 1.

Because the natural isotopic abundance of ^{13}C is only 1.108%, while that of ^{12}C is near to 99%, we would not normally observe any carbon isotope effect. Carbon isotopically

Table 1. The parameters for the three kinds of GaAs sample used.

Sample number	^{13}C doping	Resistivity ($10^7 \Omega \text{ cm}$)
1	Yes	1
2	No	1
3	No	4

**Figure 1.** The IR absorption spectrum of sample 1 (sample 3 was taken as a reference).

enriched with ^{13}C was intentionally doped into semi-insulating GaAs (sample 1) during the process of crystal growth by the LEC method. The intensity of the absorption band is defined as the product of its peak absorption coefficient α (in cm^{-1}) and the full width at half maximum (FWHM) Δ (in cm^{-1}). The concentration of ^{12}C in GaAs can be obtained conveniently from the absorption intensity and the conversion coefficient $1.1 \times 10^{16} \text{ cm}^{-2}$ [5]. Using the absorption intensity and the same conversion coefficient [6] we can obtain the concentration of ^{13}C . The actual concentrations of ^{13}C and ^{12}C measured in sample 1 are 1.4×10^{16} and $6.1 \times 10^{15} \text{ cm}^{-3}$, respectively.

In order to increase the sensitivity of the IR measurements the GaAs ingots were cut into slices that were 10 mm thick and the surfaces were lapped to a mirror finish. The infrared absorption spectra were measured using a Nicolet 7199 B-1 Fourier transform infrared spectrometer, which was operated at a resolution of 4 cm^{-1} , with 500 scans.

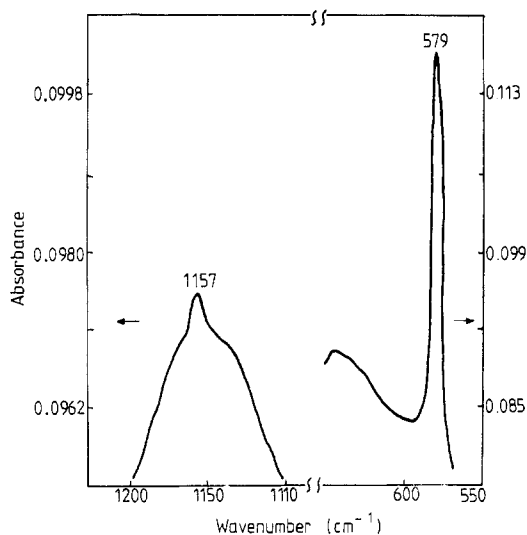
The following results were obtained.

(i) Two absorption bands at 579 and 558 cm^{-1} , corresponding to the LVM bands of $^{12}\text{C}_{\text{As}}$ and $^{13}\text{C}_{\text{As}}$ (carbon substitution on the arsenic site) respectively, were observed for sample 1 (sample 3, with the same thickness, was taken as a reference sample for the measurement) at 300 K, which is in agreement with the results of [4]. In addition, a pair of IR absorption peaks at 1115 and 1157 cm^{-1} , with exactly double the frequency of the former ones, were clearly observed, as shown in figure 1,

(ii) The absorption peaks at 579 and 1157 cm^{-1} were observed at 300 K for sample 2 without ^{13}C doping (sample 3 was again taken as a reference), as shown in figure 2. Neither the 558 cm^{-1} nor the 1115 cm^{-1} absorption band was observed for this sample. This indicates that the latter pair of bands are related to ^{13}C while the former are related to ^{12}C .

Table 2. Single-phonon and double-phonon absorption bands of $^{12}\text{C}_{\text{As}}$ and $^{13}\text{C}_{\text{As}}$ in GaAs at 300 K and 10 K.

Temperature (K)	Absorption band position (cm^{-1})			
	Single-phonon absorption		Double-phonon absorption	
	$^{12}\text{C}_{\text{As}}$	$^{13}\text{C}_{\text{As}}$	$^{12}\text{C}_{\text{As}}$	$^{13}\text{C}_{\text{As}}$
300	579	558	1157	1115
10	582	561	1164	1121

**Figure 2.** The IR absorption spectrum of sample 2 (sample 3 was taken as a reference).

(iii) In sample 1, the absorption intensity ratio of the 558 cm^{-1} band ($^{13}\text{C}_{\text{As}}$) and the 579 cm^{-1} ($^{12}\text{C}_{\text{As}}$) band is about 2.3 (indicating that the concentration ratio of ^{13}C and ^{12}C is about 2.3). Also the absorption intensity ratio of the 1115 cm^{-1} band ($^{13}\text{C}_{\text{As}}$) and 1157 cm^{-1} ($^{12}\text{C}_{\text{As}}$) is 2.4, which is consistent with the former ratio within the experimental error.

(iv) When the measurement temperature is about 300 K, the absorption intensity ratio of the 558 cm^{-1} band and its overtone at 1115 cm^{-1} is about 85 and the absorption intensity ratio of the 579 cm^{-1} band and its overtone band at 1157 cm^{-1} is about 88. These values are consistent with each other within the experimental error. The infrared absorption spectra of sample 1 were also measured at 10 K. At that temperature, both the ratios mentioned above decreased to approximately the same value—about 75.

(v) The LVM bands related to $^{12}\text{C}_{\text{As}}$ and $^{13}\text{C}_{\text{As}}$ at 300 K and 10 K are listed in table 2. Clearly the overtone relations are maintained at both 300 K and 10 K.

In view of these results, the absorption bands located at 1115 and 1157 cm^{-1} (measured at 300 K) can be unambiguously identified as the LVM two-absorption of $^{13}\text{C}_{\text{As}}$ and $^{12}\text{C}_{\text{As}}$, respectively, in GaAs.

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