Local vibrational modes of oxygen in isotopically enriched ^{28}Si, ^{29}Si, and ^{30}Si single crystals

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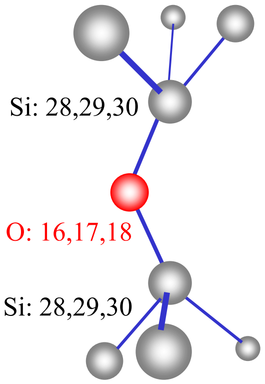
Introduction



Isotope effect in the past

Oscillating ^{16}O, ^{17}O, ^{18}O

B. Pajot, E. Artacho, C. A. J. Ammerlann and J-M. Spaeth, J.Phys: Condens. Matter 7, 7077 (1995).

D. R.Bosomworth, et.al. Proc. Roy. Soc. Lond. A. 317, 133 (1970).Hiroshi Yamada-Kaneta, Physica B, 302-303, 172 (2001)

Isotope effect in this study

Neighboring ^{28}Si ,^{29}Si, and ^{30}Si

↓

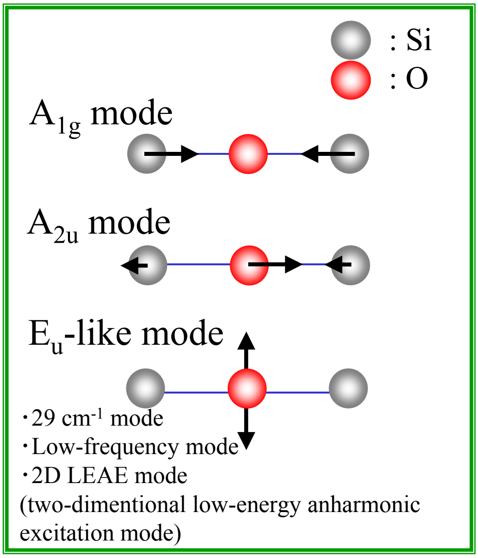
Ⅰ. LVM energy difference between ^{natural}Si(^{28}Si:92.23%), ^{28}Si ,^{29}Si, and ^{30}Si

Ⅱ. Effect of second and beyond nearest silicon

(a) LVM energy

(b) Linewidth

Local vibration modes (LVM) of O in Si



: Si

: O

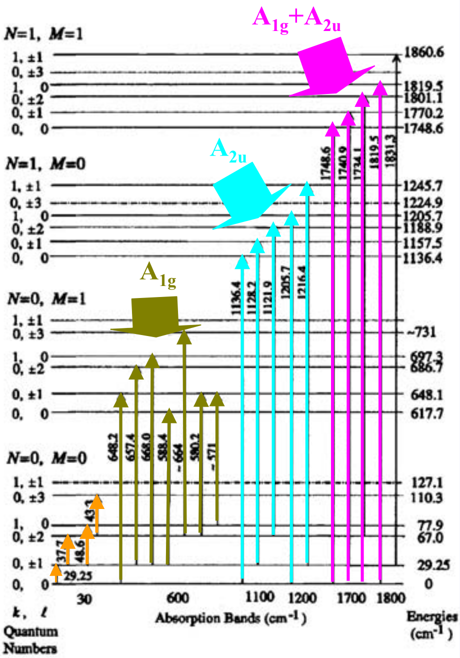
A_{1g} mode

A_{2u} mode

E_u-like mode

- 29 cm^{-1} mode
- Low-frequency mode
- 2D LEAE mode

(two-dimentional low-energy anharmonic excitation mode)



Hiroshi Yamada-Kaneta, Chioko Kaneta, and Tsutomu Ogawa, Phys. Rev. B **42**, 9650 (1990).

Samples

SI-28: CZ ^{28}Si

[O]=4.75×10^{17}cm^{-3}

(28Si:99.86%, 29Si: 0.13%, 30Si: 0.2%)

SI-29: CZ ^{29}Si

[O]=1.01×10^{18}cm^{-3}

(^{28}Si:2.17%, ^{29}Si: 97.10%, ^{30}Si: 0.73%)

SI-30: CZ ^{30}Si

[O]=8.79×10^{17}cm^{-3}

(^{28}Si:0.67%, ^{29}Si: 0.59%, ^{30}Si: 98.74%)

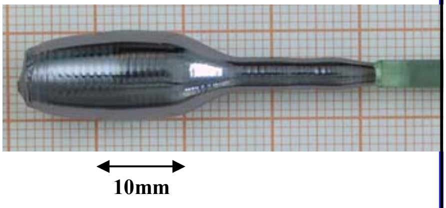
SI-Nat: CZ ^{natural}Si

[O]=6.49×10^{17}cm^{-3}

(^{28}Si:92.2%, ^{29}Si: 4.7%, ^{30}Si: 3.1%)

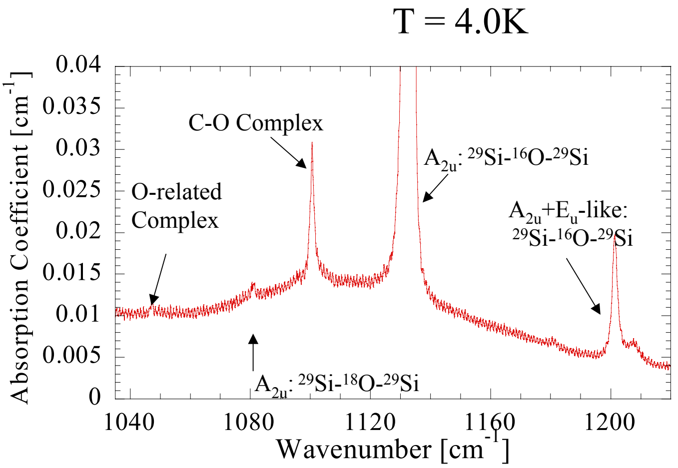
Instrument
BOMEM DA-8
Source: Globar
Detector: MCT
T=4K～290K
Resolution 0.03cm^{-1}

97% ^{29}Si single crystal

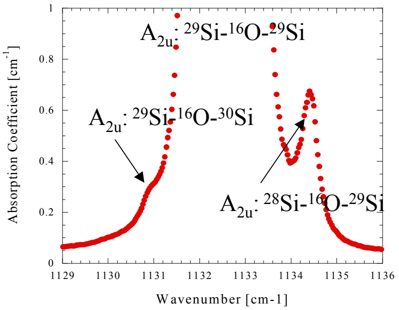


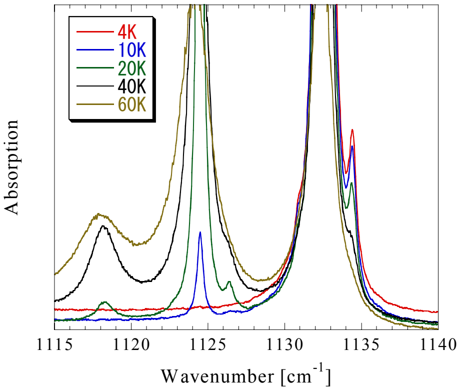
Institute of crystal growth in Berlin

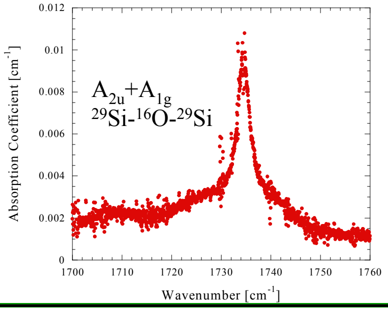
Experimental result ^{29}Si (1)



Experimental result ^{29}Si (2)

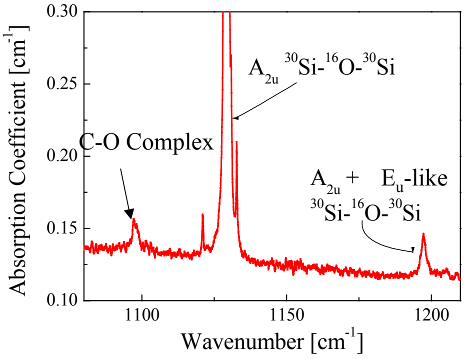


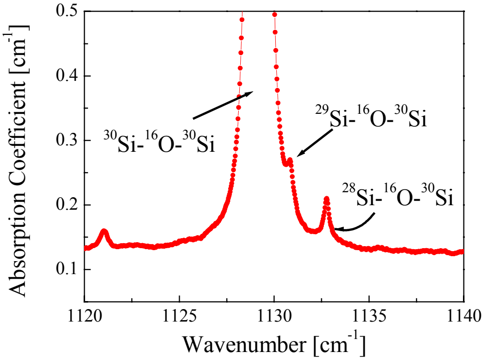




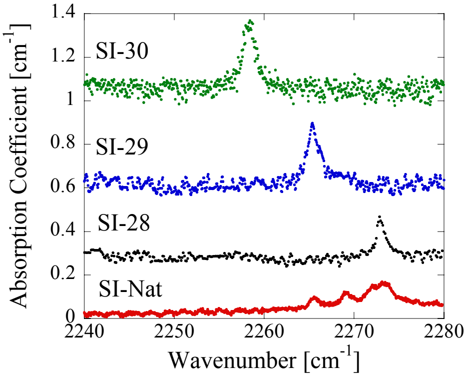
Experimental result ^{30}Si

T = 4 K

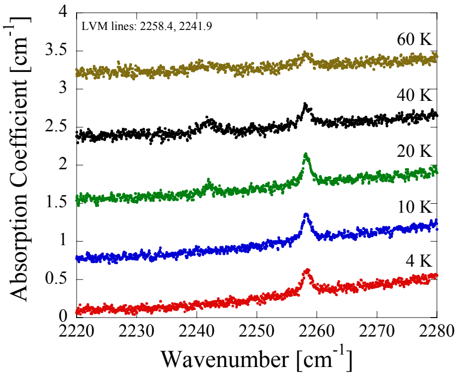




2200 lines

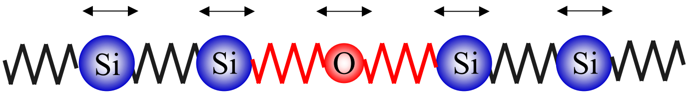


Isotope shift at 4K

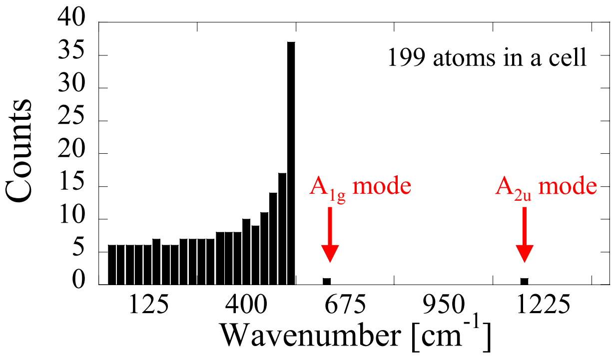


Temperature dependence of SI-30

Comparison experimental results with theoretical calculation



Bonding constant Si-Si from Raman shift of TO phonon in ^{28}Si
Bonding constant Si-O from LVM energy of A_{2u}



Comparison of experimental result and calculation

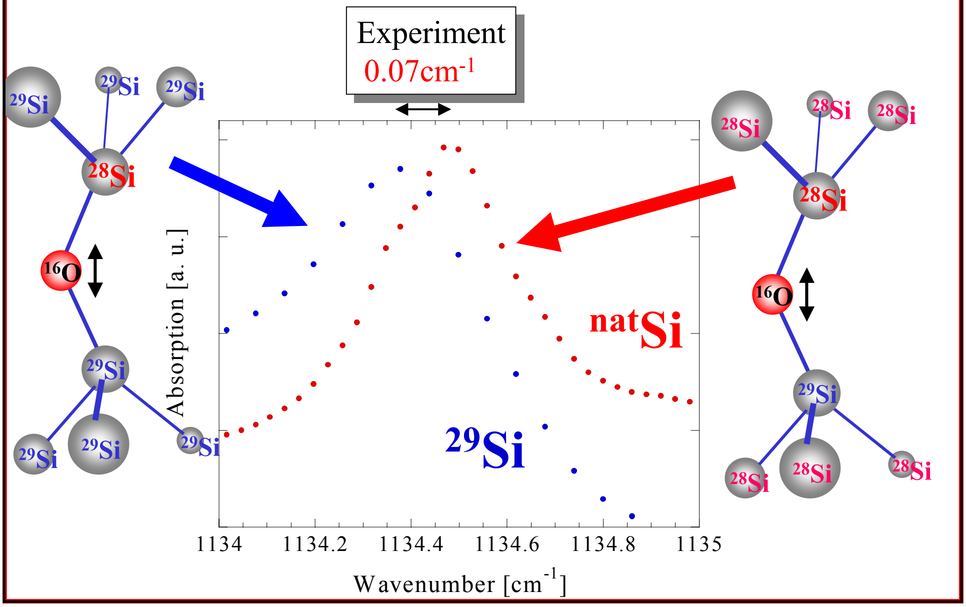
A_{2u} energy	calculation [cm^{-1}]	Experimental result [cm^{-1}]	A_{2u} energy	calculation [cm^{-1}]	Experimental result [cm^{-1}]
^{28}Si-^{16}O-^{28}Si	1136.5	1136.5	^{28}Si-^{18}O-^{28}Si	1087.4	1084.4
^{28}Si-^{16}O-^{29}Si	1134.0	1134.4	^{28}Si-^{18}O-^{29}Si	1084.9	
^{28}Si-^{16}O-^{30}Si	1131.8	1132.0	^{28}Si-^{18}O-^{30}Si	1082.5	
^{29}Si-^{16}O-^{29}Si	1131.6	1132.5	^{29}Si-^{18}O-^{29}Si	1082.2	1081.0
^{29}Si-^{16}O-^{30}Si	1129.3	1130.8	^{29}Si-^{18}O-^{30}Si	1079.8	
^{30}Si-^{16}O-^{30}Si	1127.0	1129.1	^{30}Si-^{18}O-^{30}Si	1077.4	

Positions of LVM lines

$\vert A_{2u},A_{1g},k,l\rangle$	^{28}Si ^{16}O ^{28}Si	^{28}Si ^{16}O ^{29}Si	^{28}Si ^{16}O ^{30}Si	^{29}Si ^{16}O ^{29}Si	^{29}Si ^{16}O ^{30}Si	^{30}Si ^{16}O ^{30}Si	^{28}Si ^{18}O ^{28}Si	^{28}Si ^{18}O ^{29}Si	^{28}Si ^{18}O ^{30}Si	^{29}Si ^{18}O ^{29}Si	^{29}Si ^{18}O ^{30}Si	^{30}Si ^{18}O ^{30}Si
$\vert 0,0,0,0\rangle\rightarrow\vert 0,0,0,\pm1\rangle$	29.25						27.2					
$\vert 0,0,0,\pm1\rangle\rightarrow\vert 0,0,0,\pm2\rangle$	37.7						35.3					
$\vert 0,0,0,\pm1\rangle\rightarrow\vert 0,0,1,0\rangle$	48.6											
$\vert 0,0,0,\pm2\rangle\rightarrow\vert 0,0,0,\pm3\rangle$	43.3											
$\vert 0,0,1,0\rangle\rightarrow\vert 0,1,0,\pm1\rangle$	517						517					
$\vert 0,0,0,\pm1\rangle\rightarrow\vert 0,1,1,0\rangle$	668											
$\vert 0,0,0,\pm2\rangle\rightarrow\vert 0,1,0,\pm3\rangle$	664											
$\vert 0,0,0,\pm1\rangle\rightarrow\vert 0,1,0,\pm2\rangle$	657.4											
$\vert 0,0,0,0\rangle\rightarrow\vert 0,1,0,\pm1\rangle$	648.2											
$\vert 0,0,0,\pm1\rangle\rightarrow\vert 0,1,0,0\rangle$	588.4											
$\vert 0,0,0,\pm2\rangle\rightarrow\vert 0,1,0,\pm1\rangle$	580.2											
$\vert 0,0,1,0\rangle\rightarrow\vert 0,1,0,\pm1\rangle$	570											
$\vert 0,0,0,0\rangle\rightarrow\vert 1,0,0,0\rangle$	1136.4	1134.4	1132.6	*1132.5*	*1130.8*	*1129.1*	1084.4			*1081*		
$\vert 0,0,0,\pm1\rangle\rightarrow\vert 1,0,0,\pm1\rangle$	1128.2	*1126.4*		*1124.5*		*1121.1*	1076.7					
$\vert 0,0,0,\pm2\rangle\rightarrow\vert 1,0,0,\pm2\rangle$	1121.9			*1118.3*		*1114.7*	1071					
$\vert 0,0,0,0\rangle\rightarrow\vert 1,0,1,0\rangle$	1205.7			*1201.4*		*1197.1*						
$\vert 0,0,0,\pm1\rangle\rightarrow\vert 1,0,1,\pm1\rangle$	1216.4											
$\vert 0,0,0,0\rangle\rightarrow\vert 1,1,0,\pm1\rangle$	1748.6			*1734.4*		*1721.2*	1150.8					
$\vert 0,0,0,\pm1\rangle\rightarrow\vert 1,1,0,\pm1\rangle$	1740.9			*1727.6*		*1714.2*						
$\vert 0,0,0,\pm2\rangle\rightarrow\vert 1,1,0,\pm2\rangle$	1734.1											
$\vert 0,0,0,0\rangle\rightarrow\vert 1,1,1,0\rangle$	1819.5											
$\vert 0,0,0,\pm1\rangle\rightarrow\vert 1,1,1,\pm1\rangle$	1831.3											

(in unit of cm^{-1})

Small peak shift due to host isotopes

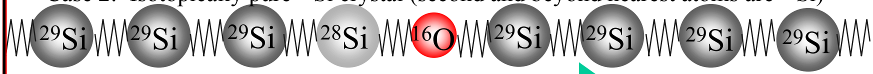


Calculation of the small peak shift

1: The effect of second and beyond nearest neighboring Si

Case 1: ${}^{natural}Si$ crystal (second and beyond nearest atoms are ${}^{28}Si$)

Case 2: Isotopically pure ${}^{29}Si$ crystal (second and beyond nearest atoms are ${}^{29}Si$)



LVM energy shift → **A_{2u} mode : 0.03cm^{-1}**

2: The effect of the lattice constant change

${}^{Nat}Si \rightarrow {}^{29}Si$: $\Delta a/a = -3.0 \times 10^{-5}$

$\longrightarrow \Delta\omega = -3\gamma \cdot \Delta a/a \cdot \omega$

γ =-0.64 (Grüneisen parameter)

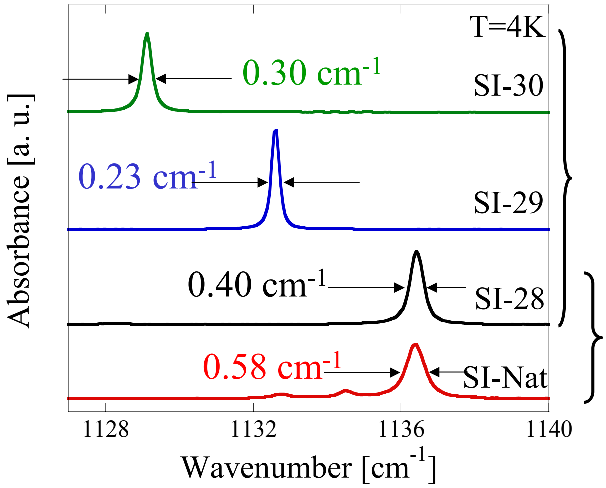
ω =1136.4cm^{-1}

LVM energy shift → **A_{2u} mode : 0.07cm^{-1}**

E. Sozontov, et al., Phys. Rev. Lett. **86**, 5329 (2001).

M. Pesola, et al., Phys. Rev. B **60**, 11449

Host Isotope effect on linewidths of LVMs



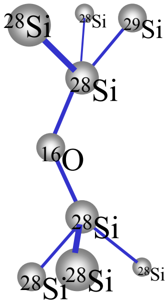
Difference of multi-phonon emission ??

Mass order effect

(Calculation ~ 0.01 cm^{-1})

Host Isotope effect on linewidths of LVMs

Calculation of mass order effect

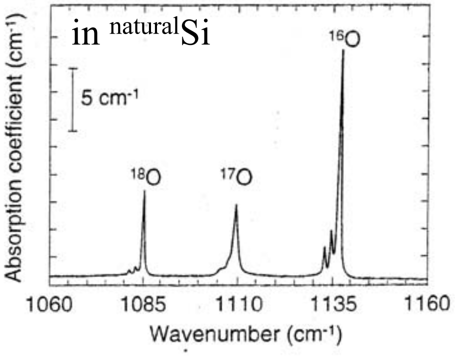


Linewidth difference between pure ^{28}Si and ^{natural}Si is

only ~ $0.01 cm^{-1}$

Host Isotope effect on linewidths of LVMs

Multi-phonon density difference



B. Pajot, E. Artacho, C. A. J. Ammerlann and J-M. Spaeth,
J.Phys: Condens. Matter 7, 7077 (1995)

Conclusion

We have measured LVMs of oxygen in isotopically enriched ^{28}Si, ^{29}Si, ^{29}Si and ^{30}Si crystals

Ⅰ. Experimentally obtained the LVM energies in ^{28}Si, ^{29}Si,^{29}Si and ^{30}Si agree with the theoretical calculation.

Ⅱ. LVM peak position also shifts due to the effect of the host Si atoms.

Ⅲ. LVM linewidth is changed due to the effect of mass disorder. However, It also depends on some other effect (one example is the difference of the multi-phonon emission).