

# Validation on the spiral microstructure formed at interface of the carbonated water in early Earth

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## [The first stage of molecular bio-history progressed by interactions between CO<sub>2</sub> and H<sub>2</sub>O]

There had existed large amount of CO<sub>2</sub> gas in the early Earth's atmosphere. The amount corresponds to 10<sup>5</sup> times of today's 0.0004 atm of CO<sub>2</sub>. The solubility in the water of CO<sub>2</sub> under the high pressure is large. However, the solubility of CO<sub>2</sub> decreases at high temperature. In carbonated water, 98.3% of the CO<sub>2</sub> is linear molecule. The linear molecule of CO<sub>2</sub> will be incorporated in vacant shaft of spiral three-dimensional structure of liquid water.

## [The lattice structure of Ice that is a model of instantaneous structure of water]

A water molecule in liquid is a tetrahedral by hydrogen bonding. The tetrahedral units are able to form various kinds of lattice structure. The lattice structure of ice is the most important model of instantaneous structure of water. The ordinary ice is symmetry of P6<sub>3</sub>/mmc (ice I<sub>h</sub>). It forms a spiral arrangement of tetrahedrons. Fig.1 shows an illustration of (ice I<sub>h</sub>) crystal basing on van der Waals radii of the atoms. There is an open 'shaft' in the center of each spiral parallel to optic axis.

## [The spiral structure that is formed by hexagonal alignment of tetrahedrons]

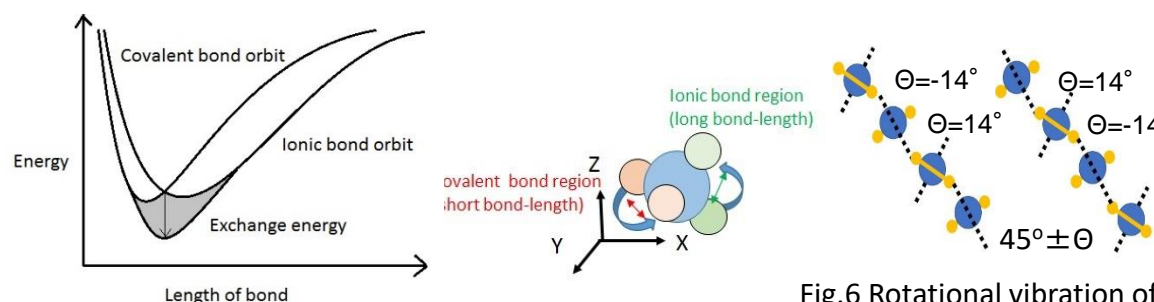
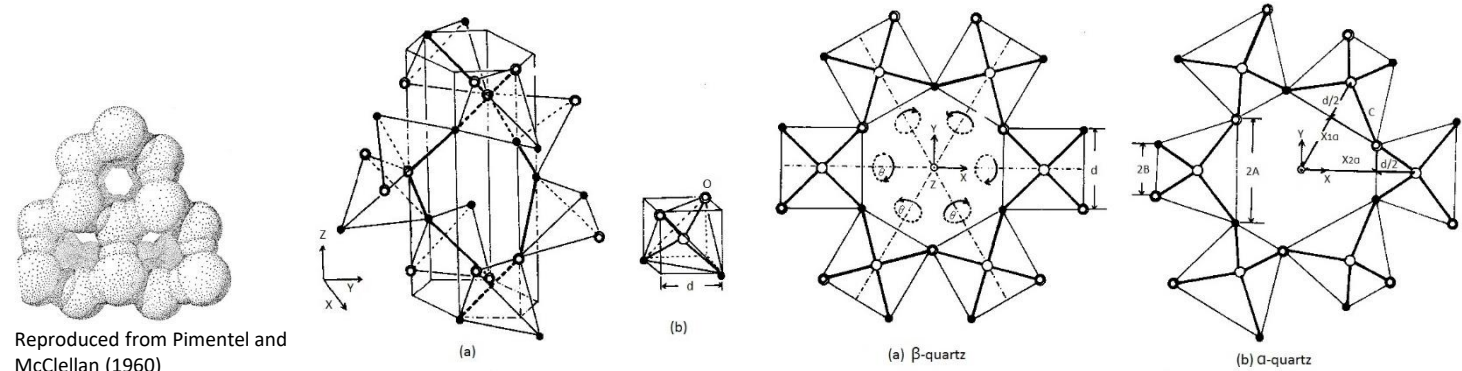
The structure on  $\beta$ -quartz is shown in Fig.2. The unit tetrahedrons are arranged helically along the optic c-(or Z-) axis. Here, one of the plane of the unit cube that contains the tetrahedron is parallel to X-Y plane. The alternative rotation of  $\theta$  (and  $-\theta$ ) on hydrogen atoms pair from 45° around its electric axis brings a geometric shrinkage of the spiral structure. Shape of trapezium in Fig.3 (b) is the projected form of tetrahedron on the X-Y plane. Where, the short side of trapezium corresponds to the side of tetrahedron that is close to the 'open' shaft vertically. The long side of trapezium corresponds to close to the X-Y plane. This rotation accompanies the larger contraction of c-axis as follows. The ratio on contraction on the X-Y plane  $[\chi(X,Y)]$  is given by Eq.(1), and that along Z direction  $[\chi(Z)]$  are given by Eq.(2),  $\chi(X,Y) = \{1+(3)^{1/2}\cos(\theta)\} / \{1+(3)^{1/2}\}$ ,  $\chi(Z) = \cos(\theta)$ ,  $\dots(1)$ ,  $\dots(2)$ . where  $\theta$  is the angle of the rotation of the tetrahedron around the electric axis. The axial ratio (c/a) for this model (Eq.(1) and Eq.(2)) is estimated as follows.  $c_{\beta c}/a_{\beta c} = 1.098$  ( $\theta=0^\circ$ )  $\dots(3)$ ,  $c_{\alpha c}/a_{\alpha c} = 1.086$  ( $\theta=14^\circ$ )  $\dots(4)$ .

## [The short-range force that deforms tetrahedron in $\alpha$ -quartz]

On the other hand, the axial ratio (c<sub>o</sub>/a<sub>o</sub>) on observed values are given by (c<sub>βo</sub>/a<sub>βo</sub>)=1.10  $\dots(5)$ , (c<sub>αo</sub>/a<sub>αo</sub>)=1.10  $\dots(6)$ . The difference between the measured value and the calculated one indicates that the model of quartz should shrink at the X-Y plane. X-ray data indicates that the silicon atom is approached to short pair of oxygen atoms slightly. The short bond length side (covalent bond side) of tetrahedron must be approached to direction of X-Y plane.

## [Flexible deformation of tetrahedral water molecule by mixing of size isomer on covalent orbit and ionic orbit]

The electronic state of water molecule in liquid is composed of covalent orbits and ionic orbits. The electronic orbital degenerated state is low energy state by the exchange interactions as shown in Fig.4. The distortion is explained by a Jahn-Teller distortion of the electronic degenerated state. The short bond length side of deformed tetrahedron corresponds to covalent region. Long bond length side corresponds to ionic bond region. The appropriate arrangement of a tetrahedron where each shaft of spiral is parallel to Z axis is shown in Fig.5.



## [Intermittent thermal vibration of respiration type in helical arrangement of water molecules]

The energy state of  $\beta$ -structure ( $\theta=0$ ) is higher than that of  $\alpha$ -structure ( $\theta>0$ , or  $\theta<0$ ). So, the torsional thermal vibration along alternating electrical axis becomes intermittently. A pair of hydrogen atoms of water molecule emerges intermittently to open-shaft of the spiral structure as shown in Fig.6. The rotation of angle ( $\theta$ ) is accompanied with a slight shrinkage. So, the vibration is accompanied with motion of breathing. Flexible deformation of each tetrahedron with the breathing motion provide the flexibility.

## [Observations of the phenomena caused by behavior of spiral structure of the water]

Followings were confirmed by slow motion replay of movie photographed at 30 frames per second [1], i.e. ① The sudden whitening of water in carbonated water was generated at the coalesce of bubbles. ② The white area of water in carbonated water behaves as if it prevents from separation of carbon dioxide bubbles. ③ Sudden movement of bubble was observed accompanying with white area in vicinity of rising bubble of carbon dioxide in carbonated water concurrently. The spiral is the lower energy structure that accompanies with a slight shrinkage. So, this whitening of carbonated water is caused by CO<sub>2</sub> incorporated in vacant shaft of spiral three-dimensional structure of the liquid water.

## [A swirling motion of electric potential at the wall of the open 'shaft' in spiral structure of carbonated water]

Most of molecules of carbon dioxide (CO<sub>2</sub>) are placed in open 'shaft' of the spiral structure. Here, electric polarization of H<sub>2</sub>O is aligned along 120-degree directional electric axis of water. The spiral structure stands along vertical direction to the X-Y plane. When thermal rotation around its electric axis is alternated ( $\pm\theta$  from 45°), the electric polarization of water molecule turns alternately. The alternately turned electric polarization in space is intermittently hanged by the thermal vibration. As shown in Fig.7, the electric potential in open 'shaft' will be moved along the spiral. The intermittent rotation of water molecule supports transportation of molecule in the open 'shaft' of spiral structure. So, instantaneous whitening of the water around the bubbles ③ is observed accompanying with the quick movement of the bubble.

## [Synthesis of hydrocarbon in the membrane that is produced by mixing of iron fine powder in carbonated water.]

When powder of iron (Fe) is mixed with carbonated water, bubbles of CO<sub>2</sub> are formed. The enlarged bubble rises up to the surface of water. The water molecules at the interface between water and CO<sub>2</sub> of gas form the spiral structure. Fine iron powder attach to open 'shaft' of each spiral in which molecules of CO<sub>2</sub> are stored. The electronegativity of carbon atom is larger than hydrogen atom. So, the iron atom (Fe) is oxidized by oxygen of CO<sub>2</sub> that is existed in open 'shaft' of spiral. The iron oxide drops down to the bottom of water. The remained carbon atom connects to a pair of hydrogen atoms in the open-shaft. A sequence of carbon atom with a pair of hydrogen atoms (-CH<sub>2</sub>-CH<sub>2</sub>-) is produced at the spiral structure. The time that the membrane of bubble becomes robust needs several hours after mixing of iron powder. carbonated water.

## [References]

[1] S. Karasawa, "The formation of spiral structure and occurrence of rotation of the spiral structure in liquid water", [ <https://www.youtube.com/watch?v=KRvJ5cCIDk> ] 2016/07/28.

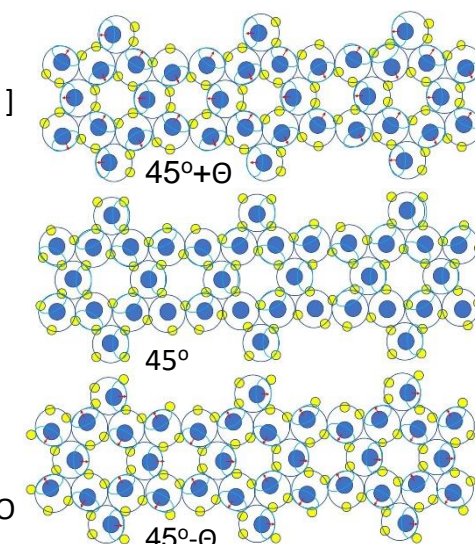


Fig.1 Top view of (ice I<sub>h</sub>) crystal. Fig.2 Lattice structure of  $\beta$ -quartz. Fig.3  $\alpha$ - $\beta$  transformation of quartz. Fig.4 The degenerated state of tetrahedral orbitals in H<sub>2</sub>O Fig.5 An arrangement of a tetrahedron Fig.6 Rotational vibration of H<sub>2</sub>O Fig.7 Swirling vibration of electric potential