Parity Violation of Electroweak Force in Phase Transitions of Single Crystals of D- and L-Alanine and Valine

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Abstract. Three kinds of experiments have been designed in attempting to observe the parity violation of electroweak force at the phase transition of single crystals of D- and L-alanine and valine. (1) An obvious λ phase transition at 270 ± 1 K was shown in the specific heat measurement of alanine and valine enantiomers by differential scanning calorimetry. The biologically dominant L-enantiomer was found to have lower energy. (2) Magnetization of single crystals of D- and L-alanine and D-valine were measured as a function of temperature using the SQUID magnetometer. The difference of the mass susceptibility $\chi_\rho \sim T$ curve between the D-alanine and L-alanine is attributable to the variation of intramolecular geometry of chirality density, which is related to the parity violation energy shift of a chiral molecule and is a consequence of the short range of the weak interaction between the nuclei and electrons. (3) Laser Raman spectra of D- and L-alanine at different low temperatures (100 K, 250 K, 260 K, 270 K, 280 K and 290 K) showed that the second order C^α -H deformation modes at 2606 cm^{-1} , 2724 cm^{-1} of D-alanine vanished at 270 K but reappeared at 100 K. In the same method, L-alanine has no such phenomenon. An obvious decrease in the scattering intensity of the methyne group C^α -H stretching mode at 2964 cm^{-1} in D-alanine occurs at the λ transition temperature. We present our experiments involving the possible relevance of Z^0 force with Salam's putative phase transition in the origin of homochirality.

Key words: D- and L-alanine and valine, parity violation of Z^0 force, phase transition

1. Introduction

In the late 1960's, S.L. Glashow, S. Weinberg and A. Salam proposed that the weak force and the electromagnetic force can be understood theoretically as different manifestations of a single underlying force: the electroweak force. According to this theory, there is a third gauge boson called the Z^0 which is neutral and mediates a new kind of weak force. Interactions involving the Z^0 gauge boson can violate parity. Since Z^0 particle is neutral, such interactions do not lead to a change in the identity of the interacting particles. Based on this mechanism, the atomic electrons

in a stable atom could be perturbed by the weak force, but in the real atom, the effects of the weak force on orbiting electrons are much too small to be detected. The three weak gauge bosons (W^+ , W^- , Z^0) predicted by the electroweak theory each have an enormous mass. It has been proven that: $m_w = (80.3 \pm 0.3)$ GeV, $m_z = (91.163 \pm 0.031)$ GeV. The mass of Z^0 particle is nearly 97.1 times as much as that of proton, 178 000 times as much as that of electron. The greater the mass of a particle, the shorter the range of the force it mediates. The photons that mediate the electromagnetic force have zero mass, and so their range is infinite. The mass of Z^0 particle is enormous, which implies that the range of the weak force is extremely short, about 2×10^{-18} meter, which is even more than 10 million times shorter than the radius of an atom. So in most orbits, electrons are actually beyond the range of weak force. To detect the effects of weak interactions, physicists have made considerable efforts, but so far, the techniques are not sensitive enough to test the electroweak force [1].

How can one make it possible to detect the effects of the weak force in the atom? If the orbit is highly eccentric or elongated, the electrons may wander close enough to the nucleus for the weak force to exert a more powerful influence.

In 1993, Salam proposed a new hypothesis [2]: the subtle energy difference of chiral molecules induced by Z^0 interactions combined with Bose condensation may cause biochirality among 20 amino acids which make up proteins to be a consequence of second order phase transition below a critical temperature T_c , which is analogous to that of BCS superconductivity. The T_c for BCS superconductivity for metals is of the form $\omega \exp(-2/g_{eff}\sigma(0))$, Salam conjectured that a similar formula may hold for the case of amino acids chains. A crucial form for the transition temperature T_c involves dynamical symmetry breaking. The t -quarks or supersymmetry appear to be essential if such mechanisms are to hold. To deduce the value of temperature T_c , one is supposed to start from the Feynman Lagrangian methodology of the BCS theory for the superconducting electronic system and tries to write down the equivalent Ginzburg-Landau equation, from which the following result is presented:

$$T_c = \frac{\langle \varphi \rangle}{10^3} \exp(-2/g_{eff}\sigma(1 - 4\sin^2\theta)) \approx 2.5 \times 10^2 \text{ K}.$$

Here φ field is expressed as a complex auxiliary Higgs scalar field. In terms of the quantity ($\langle \varphi \rangle^0 = 250$ GeV) the electron mass M_e turns out to be a very tiny number $\approx 2 \times 10^{-6}$. Such a number is large only for the top quark if its mass is in excess of 100 GeV. φ is considered as a $t\bar{t}$ composite.

g_{eff} is an attractive coupling constant between spin up ($\uparrow$) and spin down ($\downarrow$) electrons. To compute further, it must be guaranteed the g_{eff} in the effective Lagrangian is a positive number corresponding to attraction. Assuming that $\sigma(0) \approx m_z^2$, we obtain $g_{eff}\sigma(0) \approx 1$; it was taken $(1 - 4\sin^2\theta) \approx 1/13$ with the empirical value of the parameter $\sin^2\theta \approx 0.231$, then the exponential factor gives $\exp(-26) \approx 10^{-10}$.

Table I. C, H, N element analysis of D- and L-alanine

element	sample alanine	experimental percentage %	theoretical percentage % $\text{C}_3\text{H}_7\text{O}_2\text{N}$ ($M = 89.11$)	relative error %	theoretical* percentage % $\text{C}_3\text{H}_7\text{NO}_2 \cdot \text{H}_2\text{O}$ ($M = 107.13$)
N	D	15.96	15.72	1.5	13.08
	L	15.75	15.72	0.2	13.08
C	D	40.33	40.43	0.24	33.63
	L	40.49	40.43	0.15	33.63
H	D	7.88	7.93	0.76	8.49
	L	7.77	7.93	2.01	8.49
O	D	35.83	35.92	0.25	44.80
	L	35.99	35.92	0.2	44.80

When we attempt to lay the experimental bases of the origin of homochirality, some fundamental questions arise. Whether the putative second order phase transition of the type of Bose condensation may occur in amino acids? The range of the neutral-weak force is extremely short, how can we get some information about relatively long-range effects of the Z^0 force? The putative phase transition may induce molecular changes that lead to the breaking of chemical bonds. It still suffers from the difficulty in explaining of overcoming the large activation energies to break the chemical bonds for a given D amino acid to be converted to L type. Three kinds of experiments were carried out to observe the putative phase transition and the possible role of electroweak force at the phase transition of single crystals of D- and L-alanine and valine.

2. Materials and Methods

The powder of D- and L-alanine samples were obtained from Sigma Chemical Co-operation. The single crystals, which were grown by slow evaporation of saturated aqueous solutions at 4 °C, then washed with absolute alcohol, evacuated and kept in a desiccater, were well-formed crystals elongated along the c axis.

2.1. CHARACTERIZATION OF SAMPLES

2.1.1. Element analysis

Element analysis of C, H, N of D- and L-alanine was carried out (Table 1). The powder was prepared from the crystal samples.

It's shown in Table 1 that there's a good agreement between the theoretical and experimental data. The relative error is within the limits of experimental error of

Table II. Crystallographic data for crystals of D- and L-alanine

Formula	C ₃ H ₇ NO ₂ (D-form)	C ₃ H ₇ NO ₂ (L-form)
Formula weight	89.10	89.10
Crystal size (mm ³)	0.40 × 0.25 × 0.20	0.40 × 0.35 × 0.30
Crystal system	Orthohombic	Orthohombic
Space group	<i>P</i> 2 ₁ 2 ₁ 2 ₁	<i>P</i> 2 ₁ 2 ₁ 2 ₁
<i>a</i> (Å)	5.7840(10)	5.7840(10)
<i>b</i> (Å)	6.0250(10)	6.0250(10)
<i>c</i> (Å)	12.331(2)	12.331(2)
<i>V</i> (Å ³)	429.72 (12)	429.72 (12)
<i>Z</i>	4	4
<i>D</i> _{calc} (g cm ^{−3})	1.377	1.377
Absorption coefficient (μ)	0.115 mm ^{−1}	0.115 mm ^{−1}
<i>F</i> (000)	192	192
Scan mode	2θ − θ	2θ − θ
2θ range(°)	3.30–24.98	3.30–24.95
No. of reflections	469	463
No. of unique reflections	469	463
<i>R</i>	0.0346	0.0336
<i>R</i> _w	0.0367	0.0340

3%. Furthermore, to suppose there is a crystal water, comparing the theoretical percentage in the sixth column with the actual experimental value, we exclude the possibility of the existence of water in the crystal samples.

2.1.2. X ray single crystal diffraction

The data for D-/L-alanine single crystals were collected at room temperature on a Siemens R3m/V diffractometer with Mo-K_α (λ = 0.71073 Å) radiation. Crystal data and data collection parameters are listed in Table 2. The structure was solved by direct methods and subsequent Fourier differential techniques and refined by full-matrix least squares using the SHELXTL PLUS program. All non-hydrogen atoms we refined with anisotropic thermal parameters. By using X-ray crystallography, the cell dimensions of D-/L-alanine crystals were determined as the following: space group *P*2₁2₁2₁, *a* = 5.7840, *b* = 6.0250, *c* = 12.331 Å, *V* = 429.72 Å³, which agrees quite well with the reported literature [3]. The X-ray structure determination indicates that the D-/L-samples are pure single crystals, containing no crystal water.

Table III. Rotation power of D- and L-alanine

sample	ζ	calculated α	reference	relative error
D	-0.09	-1.76	-1.8	2%
L	0.153	1.78	1.8	1%

$[\alpha]$: the rotatory power

L: the path length (1 dm)

ζ : rotation angle

C: concentration of solutions of D-Ala and L-Ala is 0.051, 0.86 (g ml⁻¹)

2.1.3. Rotatory power measurement

Rotation power at a certain temperature and wavelength ($[\alpha]_{\lambda}^T$) is one of the basic characteristics of optically active materials. To further determine the purity of our samples, we measured on Polarimeter PE-241MC the rotation angle ζ of D- and L-alanine solutions respectively, at 19.5 °C and the wavelength of 589.6 nm, then by using the formula of $[\alpha] = \zeta/(L \times C)$, we got the corresponding α of D- and L-alanine.

Based on the experimental results, the deviation between the experimental result and the theoretically value is definitely within the experimental error, which indicates that no other optically active impurities exist.

Additionally, the water content in all samples before and after cooling was detected to be less than 10 ng by Infra Photoacoustic Spectra. Taking these two factors into consideration, a conclusion can be drawn that D- and L-alanine single crystals are exactly enantiomers without water molecule in each crystal.

2.2. SPECIFIC HEAT MEASUREMENTS

We designed the first experiment to measure the specific heat by differential scanning calorimetry (DSC) with an adiabatic continuous heating method. The typical size of the single crystals used in DSC were $5.0 \times 5.0 \times 1.0$ mm³. The experiments were performed on D-alanine (sample 1: 113.30 mg), L-alanine (sample 2: 99.40 mg), D-valine (sample 3: 67.47 mg) and L-valine (sample 4: 40.20 mg) [4].

For the purpose to compare the energy difference between D- and L-valine at T_c on the base of theoretical calculation [6], sample 3 (D-valine, 30.28 mg) was mounted on the sample holder and sample 4 (L-valine, 30.25 mg) as the reference sample, we obtained data for the differential specific heat between D-valine and L-valine in the same system with exactly the same condition (Figure 1).

The differential specific heat was measured by increasing temperature with the temperature scanning speed set to 0.5 K min⁻¹ in the region of 77 ~ 300 K. The temperature accuracy was better than ± 1 K at the measured temperature range.

Figure 1. Temperature dependence of the differential specific heat between D-valine and L-valine.

2.3. DC MAGNETIZATION MEASUREMENTS

The second experiment was performed by applying an external magnetic field to the atoms of the molecular crystals of D- and L-alanine, with the aim to get some information about the effects of weak force. The external magnetic field was implied to provide the atoms in the molecule a certain preferred direction of spin. We measured magnetic moments (m) and magnetic susceptibilities (χ_ρ) of D- and L-alanine single crystals by a DC MPMS-5 SQUID magnetometer at a field of 0.1 T in the temperature ranging from 200 K to 300 K. Because the magnetization change per sample was close to the baseline resolution of the magnetometer, care was needed to design the sample holder. The sample was held directly in the center of a straight piece of plastic straw with a diameter close to that of the sample crystals. Data points are spaced by 3 K. Different magnetic behaviours of D- and L-alanine as well as D-valine molecules have been observed in our experiments, as is shown in Figure 2. Data for L-valine are absent due to the difficulty to obtain a big enough single crystal.

We further carried out a similar measurement in a field of 1 T. To improve the relative accuracy, we used a stronger field and bigger samples, the magnitude of the magnetic moment is on the order of 10^{-3} emu, 10^2 times stronger than those in a field of 0.1 T. The same result was shown from the data (Figure 3).

Figure 2. Temperature dependence of mass susceptibility (χ_ρ) $H=0.1$ T. (a) L-alanine; (b) D-alanine; (c) D-valine.

Figure 3. Temperature dependence of mass susceptibility (χ_ρ) $H = 1$ T. (a) D-alanine; (b) L-alanine.

2.4. RAMAN SPECTRAL MEASUREMENTS

From the configurations of the enantiomers of amino acids, we can see that, to change the configuration of an amino acid from D to L, it seems necessary to alter a bit for the vibration modes of the $C^\alpha-H$ to facilitate the transition. So, in Raman spectra measurement, we put our emphasis mainly on the observation of the possible alterations of $C^\alpha-H$ bond vibration under the variation of low temperatures.

Figure 4. Raman spectra of D- and L-alanine single crystals at different temperatures.

Based on the above rules, we designed the third experiment to measure the laser Raman spectra of D- and L-alanine single crystals using a SPEX 1403 double-grating monochromator with a C 31034 photomultiplier tube. The 488 nm line of an Ar ion laser was employed as the excitation source with an incident power of 200 mW. A band filter for the incident laser was inserted into the optical system. The incident laser beam was along the c axis and polarized along the b axis, the emissions were collected along the a axis, focused into a Spex 1403 double monochromator and detected by a photomultiplier followed by a Spex-datamate. Data acquisition times of between $2 \sim 4$ s/data point were employed and data points are spaced by 2 cm^{-1} (Figure 4).

3. Results and Discussion

3.1. SPECIFIC HEAT ANALYSIS

In this measurement, an obvious λ transition is observed at $270 \pm 1 \text{ K}$ (see Figure 1 and Figure 2 in Reference [4]). The phase transition is reversible. Although Grunenberg A. et al. [5] studied 22 crystalline aliphatic amino acids including L- and D-valine by DSC in the temperature ranging from 233 to 423 K, such kind a transition was not observed in the single crystals of valine.

With all the other conditions being equal, it is also shown that the specific heat C_p value of D-valine is larger than that of L-valine and the same existed in the enantiomers of alanine. In all cases, the biologically dominant L-enantiomer is found to have lower energy, and the specific heat value reflects this fact.

In Figure 1, we got a differential peak, which shows 10^{-4} eV per molecule difference between D- and L-valine at the transition point. It has been 10^{15} times greater than the theoretical energy difference of $3 \times 10^{-19} \text{ eV/molecule}$ [6]. According to the formula $2\Delta(0) = 3.53k_B T_c$, the electron coupling energy is on the order of $10^{-3} \sim 10^{-4} \text{ eV}$ per molecule, which coincides well with the measured energy difference in λ peak of D-valine using L-valine as reference. We propose that the specific heat jump of D-valine may be attributed by the electron coupling to form Cooper pair. The free electrons of H atom tend to change to superconductive electrons.

Theoretically, the specific heat of crystal is influenced by the lattice and electrons structure. However, the result of X ray powder diffraction demonstrated that the effect of crystal lattice on C_p could be excluded and the phase transition observed might be ascribed to the contribution of electrons structure.

3.2. MASS SUSCEPTIBILITY ANALYSIS

During the measurements, we got $m \sim T$ curves at different temperatures. The relationship between the magnetic moment and mass susceptibility can be expressed as the following formula:

$$\chi_\rho = m/(H \times g)$$

in which χ_ρ is mass susceptibility ($\text{cm}^3 \text{g}^{-1}$), m is the magnetic moment (emu), H is the magnetic field (G), g stands for the mass of each sample. Based on this formula, we transferred the $m \sim T$ curves into the corresponding $\chi_\rho \sim T$ curves shown in Figure 3.

As known from Figure 3, weak diamagnetisms were observed in two cases. Valine and alanine enantiomers are diamagnetic, since they have even numbers of electrons which form closed, magnetically neutral shells.

Their magnetic susceptibilities are negative ($\chi_\rho < 0$), the values are very low and in absolute value about $10^{-7} \text{ cm}^3 \text{g}^{-1}$. In Figure 2(a), 2(b) and (2c) we present the temperature dependence of the susceptibilities of D- and L-alanine and D-valine from 200 to 290 K and find that the temperature at which the discontinuity of magnetic susceptibility occurs for D-alanine is coincident with T_c of specific heat measurement. It is obvious that the χ_ρ vs T is different for D- and L-alanine.

The ordinary parity-conserving electromagnetic forces between the electrons and the nuclei in the molecule tend to align the axis of each electron's orbit against its axis of spin. This phenomenon is referred to as spin-orbit coupling. For a right-handed helical molecule, spin-orbit coupling favors down spiraling for spin-up electrons and up spiraling for spin-down electrons. In either case the spin axis of the electron tends to be aligned against the electron's direction of motion, so that in a molecule shaped as a right-handed helix, spin-orbit coupling produces predominantly left handed electrons, while in left-handed helix, right-handed electrons predominate. As a result, molecules display regions of different electron chirality [7]. Thus, different magnetic behaviour for D- and L-amino acids has been observed in the experiments.

It is also interesting to notice that in both D-alanine and D-valine, unlike the corresponding L-cases, there is a jump in the $\chi_\rho \sim T$ curve around 270 K, leading to a maximum of χ_ρ . It's the same with the second measurement in a field of 1 T, which strengthened our former experimental results (Figure 3).

Now we can switch on the Z force. The difference of the $\chi_\rho \sim T$ curve for D-alanine and L-alanine single crystals might be attributed to the different interaction of Z force with the right-handed electrons and left-handed electrons, it produces an energy shift in the molecules: the energy of one enantiomer (D-form) is increased and that of the other (L-form) is decreased [8]. Quantum mechanical theory indicates that the magnetic susceptibility of a system with weakly interacting particles, for the case where the electron clouds of the particle are not spherically symmetrical, will be composed of two parts: (A) precession diamagnetism (B) Van Vleck polarization paramagnetism. At elevated temperature, the role of polarization paramagnetism will be greater (in the D-case), while in the L-case, the orientation paramagnetism which decreases with temperature predominantly affects the magnetism [9]. The coupling of the variation of intramolecular geometry of chirality density and the weak interaction between the nuclei and electrons caused the discontinuity in susceptibility.

3.3. RAMAN SPECTRA ANALYSIS

The results of Raman spectra (ranging from 100 K to 290 K) of the single crystals of D- and L-alanine are shown in Figure 4.

For the special purpose to study the mechanism of the observed transition, we concentrated on the spectral region concerning various modes of the C* (C $^{\alpha}$)-H bond, since the transition of D- to L-amino acids usually requires the breaking of α C-H bond. Within the region of 2500–3100 cm $^{-1}$, there are seven peaks at 2606(m), 2724(m), 2888(m), 2934(m), 2964(s), 2988(s), 2998(s) cm $^{-1}$ respectively, the former two peaks are assigned to second order C $^{\alpha}$ -H deformation, the other five peaks were assigned to C-H stretching, among which, one intense band at 2969 cm $^{-1}$ is due to the stretching mode of the methyne group, ν (C $^{\alpha}$ -H). By comparison with well-known methyl group frequencies [10], of the five medium-to-strong high frequency Raman bands observed, three other Raman bands at 2934(m), 2988(s), 2998(s) [11] are indicated to the three CH $_3$ stretching modes. The remaining medium band at 2888(m) is likely the overtone 2δ (CH $_3$) in Fermi resonance with the symmetric CH $_3$ stretch at 2932 cm $^{-1}$.

The spectral intensity distribution is asymmetric, with higher background at low frequency side. The asymmetry enhances as temperature is lowered, resulting in λ shape with a sharp peak, which indicates that the C $^{\alpha}$ -H stretching modes are anharmonic. As shown from Figure 4, although the spectral shape and width of most peaks remain almost constant for temperature varied from 100 to 297 K, the C $^{\alpha}$ -H deformation modes of D-alanine at 2606, 2724 cm $^{-1}$, weak and broad, vanished at 270 K. We studied temperature dependence of the integrated intensity for these two peaks, as is shown in Figure 5. A large decrease in the scattering intensity of the two peaks occurred when temperature was changed from 297 K to 270 K; while temperature was changed from 270 K to 100 K, the intensity picked up and could be resolved again, but L-alanine has no such phenomenon. Another noticeable difference between spectra of D- and L-alanine exists between the activities of peak 2964 cm $^{-1}$, which is assigned to be C*-H stretching. The integrated intensity at different temperatures for this peak (as shown in Figure 5), underwent a change similar to that of 2612, 2728 cm $^{-1}$, but it didn't disappear at 270 K.

To provide certain illustration, we take the simplified lattice structure of D- and L-alanine as explanation. The alanine crystals are orthorhombic with space group $P2_12_12_1$. Figure for the bond distances and angles of L-alanine single crystal is shown on the right. The left figure presents a view down the C(2)-C(1) bond, showing conformation of the heavy atoms [3].

The α -proton left behind migrates to the nitrogen so that the Z^0 meson before being annihilated gives rise to an interaction between the quarks contained inside the proton and inside the nitrogen [2]. The Z^0 component of the electroweak force might transfer the parity violation from the particle one to the molecular one, giving rise to a phase transition. The Z^0 force interacts with D- and L-alanine in different

Figure 5. Relative intensity of the three C^α -H modes vs T (K).

Figure 6. Crystal structure of D- and L-alanine

ways. The temperature dependence curves of relative intensity of the two second order C^α -H deformation modes are supportive to the above-mentioned discussion.

4. Conclusion

For a long time, classical chemists have ignored the effect of the electroweak interaction, due to the predominance of electromagnetic force in chemical effects. It seems impossible for so small a 'parity-violating energy difference' between the enantiomers to lead to a phase transition of D- to L-amino acids, as proposed by Salam. However, the Z^0 component, in spite of the fact that its effects appear to be negligible at low energies, may play an active role in homochirality. We will show that the electroweak interactions (for which there is nothing comparable in classical chemistry) can give rise to a phase transition.

Like all phase transitions, this one will have a behaviour like $(T_c - T)^\alpha$, $T \rightarrow T_c$. This makes the amplitude an infinite quantity. As to the question of the effects of the parity violating weak interactions for specific amino acids, it can be answered by setting [12] $(1 - 4 \sin^2 \theta) \approx 0$, the resulting expression for the Hamiltonian has led to *molecular-conformation-dependent* values for Parity violation which for Ala, Val, Ser and Asp have been calculated to give -3.0 , -6.2 , -2.3 , -4.8 in units of 10^{-19} eV [6]. To compute T_c exactly is difficult on account of the incompleteness of the standard model of elementary particles, but we can measure T_c by using equipments with high sensitivity and accuracy.

The mutual support of the above three different kinds of experiments, in reaching a possible conclusion, illustrates the effect of the parity violation of electroweak force induced anomalous behavior at the phase transition point of D- and L-alanine and valine. The cooperative phenomena in condensed crystals leads to our suggestion that the Z^0 force induced energy difference can be amplified at low temperature at the transition point.

Further in-depth research for the underlying factor is in progress.

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