Measurements of the Diamagnetic Susceptibility of NaCl Aqueous Solution

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Abstract: Using a SQUID magnetometer, the diamagnetic susceptibility of NaCl aqueous solution was measured with high accuracy in a magnetic field of up to 6 T at 25±0.05°C. The NaCl concentrations adopted in this experiment were 0 (water), 7.5, 15, 23, 26.2, 26.6 and 100% (crystal) with the concentration error of ±0.04%. Experimental data was compared with the calculated value of susceptibility derived from dependence of the vapor pressure on NaCl concentration. As a result, our measured value was almost in accordance with the calculated value. It was found that the diamagnetic susceptibility shows a decrease of approximately 10% within the saturated concentration (26.2%) and that the susceptibility is one of the effective cause for the concentration dependence in the gas-liquid interface deformation of the NaCl solution.

Key words: Concentration dependence, Magnetic susceptibility, NaCl aqueous solution, Diamagnetism

1. Introduction

The gas-liquid interface of NaCl aqueous solution with diamagnetism begin to cave downward at the magnetic center by applying magnetic field. This is because the pressure of the interface at the magnetic center decreases and the gas-liquid interface is stretched to both ends by magnetic forces. It was found that the gas-liquid interface deformation increased in proportion to the square of magnetic field and that the deformation gradually diminished with increasing NaCl concentration (S-H, 2001). Especially, the latter showed a decrease of approximately 50% against the increase of 20% concentration at 6 T. It is clarified that this is related to the diamagnetic susceptibility and density depended on the NaCl concentration. Accordingly, the accurate data of the susceptibility and the density of the NaCl solution over a wide concentration range should be required in order to carry out the quantitative analysis of the gas-liquid interface deformation. However, the data satisfying NaCl concentration dependence of the susceptibility in comparison with the density has not been obtained.

Moreover, the susceptibility of NaCl solution is an important physical parameter in studying the application of magnetic fields to seawater, e.g., electromagnetic propulsion and ocean current magnetohydrodynamic (MHD) power generators, and the magnetic influence acting on the biological system as well as on the growth of microorganisms in a culture solution, because a large number of Na⁺·Cl⁻ ions are included in all these research objects and NaCl concentration is thus a major factor. The aim of the present study is to measure the susceptibility of NaCl solution carefully by means of the latest measurement method over a wide concentration range in order to elucidate a cause of the concentration dependence in the gas-liquid interface deformation of the solution.

2. Experimental Setup

A superconducting quantum interference device (SQUID) magnetometer (Quantum Design, MPMS-7) was used in the measurement of susceptibilities of water and NaCl solutions. The maximum magnetic field of this magnetometer is 7 T and the homogeneity of the magnetic field in the sample space is ±0.1%. Figure 1 shows a schematic diagram of the experimental apparatus and sample cell. As a sample cell, a Pyrex glass ampoule with the outer diameter of 7.0 mm, the thickness of 1.0 mm, the height of 17.5 mm, and the caliber of 0.8 mm was used. The sample solution was made by dissolving NaCl crystals (with the purity of 99.5%) in distilled water (with the electric conductivity of 1.0×10⁻⁶ S/m) and was prepared at seven different NaCl concentrations of 0 (water), 7.5, 15, 23, 26.2, 26.6 and 100% (crystal) with the concentration error of ±0.04%. NaCl 26.2% was included in the object of the measurement to investigate the behavior in the vicinity of the concentration of a saturated solution, and NaCl 26.6% higher than the saturated concentration was included to examine the change of the susceptibility at the concentration at which the crystals began to precipitate. Each ampoule cell was filled with the sample solution of

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![Schematic diagram of the experimental apparatus and sample cell.](image)

Fig. 1 Schematic diagram of the experimental apparatus and sample cell.

117.85-155.10 mg (±0.05 mg) by syringing each 100 ml mother liquor swung sufficiently at 25°C and was covered tightly. It was noted that the temperature of the solution of NaCl 26.6% was sufficiently raised and then the solution was injected into the ampoule cell in the condition without the crystal, because the solution was over the concentration of saturated solution (Handbook, 2000) (26.45% at 25°C). The tightening was carried out by the method of instantaneous welding the tip of the ampoule cell with the oxygen gas burner; after the sample injected the solution was rapidly cooled under the freezing point. By using this method, it was possible that there was seldom the change of the mass of the whole sample after the welding (maximum mass loss of under -0.05 mg). Also, the tightening was confirmed through the mass measurement after vacuum pumping (0.3 Pa). The influence of susceptibility of the air being in the top part of the sample cell in Fig. 1 was disregarded because it was three orders of magnitude smaller than the susceptibility of NaCl solution by calculation.

In this experiment, the sample was connected to the sample probe for the measurement using an exclusive contracting tube. The sample probe was set so that the sample was at an optimum position with respect to the sensing coil in a Dewar vessel. Then, the sample space was maintained at 25°C, and the magnetization of the sample was measured in the magnetic field up to 6 T. When the measurement was completed, the sample was taken out from the magnetometer to observe the reproducibility of the measured magnetization, and the measurement process mentioned above was repeated several times. In addition, it was confirmed that there was no mass change of the sample throughout the magnetization measurement process.

![Graph showing magnetization vs. magnetic field for different NaCl solutions.](image)

Fig. 2 Average magnetization of water and NaCl solutions.

3. Experimental Results

3.1 Magnetization of sample solutions

The measurement result of magnetization at 25±0.05°C for the seven sample solutions of NaCl concentrations is shown in Fig. 2. The magnetization of the glass ampoule cell has been canceled in the measurement. This figure depicts average magnetizations of water and five NaCl solutions in the magnetic field range of 0–6 T. In Fig. 2, each magnetization curve is linear in the range of the applied magnetic field. When the magnetization measurement of the water was performed three times, the reproducibility of the measurement results was excellent. Also, there was a good reproducibility for the magnetization of NaCl solutions in the measurements repeated three times (six times on NaCl 15 and 23%). It is proven that the gradients of these magnetization curves, namely, the susceptibility, decrease in the order of water, NaCl solution under the saturated concentration (7.5–26.2%) and NaCl solution over the saturated concentration (26.6%).

Figure 3 shows susceptibilities of water and NaCl solutions. The susceptibility was obtained from the average magnetization in Fig. 2. The solid line connects the susceptibility for each NaCl concentration. In Fig. 3, the susceptibility curves are straight lines which are parallel to the horizontal axis. This result shows that susceptibilities of water and NaCl solutions do not depend on the magnetic field in the range of 2 T to 6 T and their magnetizations do not reach the magnetic saturation condition in the field up to 6 T.
3.2 Concentration dependence of susceptibility

The relationship between the susceptibility and NaCl concentration in the sample solution was examined in detail. The susceptibility of the sample solution was obtained from the gradient of the measured magnetization curve by the least squares method. The result is shown in Fig. 4. The horizontal axis is NaCl concentration and the vertical axis is susceptibility in this figure. The solid circles indicate the measured value, and they are connected by a solid line for each NaCl concentration. The open circles denote the susceptibility of water and NaCl crystal reported in the CRC(Handbook, 2000), and the open triangles depict the susceptibility of NaCl crystal and NaCl solution near the saturated concentration as reported by Hocart(R. Hocart, 1929). NaCl concentration of the saturated solution (26.4%) at 25°C is shown by the dotted line. Each measured value was appended to the error bar by the mean square error.

Based on Fig. 4, the measured value for susceptibilities of water and NaCl crystal was compared with the values obtained in the past by other researchers. The measured value of the susceptibility of water was $-6.67 \times 10^{-7}$ emu/g which was 6.9% lower than the value obtained in the past. The susceptibility of water $-7.20 \times 10^{-7}$ emu/g as a standard at 25°C was used. This value was obtained by substituting the susceptibility(Handbook, 2000) at 20°C for the empirical formula of Philo–Fairbank(J.S., 1980) obtained in a magnetic field of 0.43 T. Since the reference value which had a high reliability for susceptibility of water was not determined(J.S., 1980) (R. Cini, 1968), our measured value should be used as the reference susceptibility of water. On the other hand, the measured susceptibility of NaCl crystal was $-4.85 \times 10^{-7}$ emu/g which was 6.2% smaller than the value(Handbook, 2000) $-5.17 \times 10^{-7}$ emu/g obtained previously. This difference was considered to be caused by impurities (K, Mg, Ca, Ba etc.) of 0.5% contained in NaCl crystal (99.5% purity) which weakened the diamagnetism of NaCl.

The susceptibility between water and NaCl solution was compared and the concentration dependence on the susceptibility was discussed. The measured susceptibility of NaCl solutions of 0 (water), 7.5, 15, 23 and 26.2% were $-6.67$, $-6.29$, $-6.10$, $-6.01$ and $-6.06 \times 10^{-7}$ emu/g, respectively. It is evident from Fig. 4 that the susceptibility of NaCl solution in any case is clearly less than that of water. Also, the susceptibility decreased gently with increasing NaCl concentration from 0-26.2% (concentration of saturated solution). The susceptibility of these NaCl solutions showed a small decrease of 9.9% within concentration of saturated solution (26.2%). In addition, the susceptibility was rapidly reduced to $-5.28 \times 10^{-7}$ emu/g at NaCl 26.6% which exceeded the saturated concentration. Compared with the value(R. Hocart, 1929) of NaCl 22.1-26.5% obtained by Hocart, the measured values of NaCl 23% and 26.2%, were 13.6-15.6% greater. The cause of this difference is discussed as follows. Hocart measured the force working on NaCl solution in a gradient magnetic field by the level raising method (the Quincke method) and the pendulum method of Weiss–Föex–Forrer(R. Hocart, 1929), and the susceptibility was obtained from the force in comparison with that of water at 20°C. However, the magnetic field used by these measurement methods was much smaller than 6 T used by us. It follows that, the intensity of the magnetic signal received in his measurement was also considerably weak.
hence it is considered to be caused by the large measurement error included. The calculated value for susceptibility of NaCl solution should be obtained in order to compare it with our measured value as the next step.

4. Discussion

The data of susceptibility was quantitatively analyzed on the basis of the relationship between NaCl concentration and interaction force between water molecules and ions. It is known that the diamagnetic susceptibility increases with the temperature due to weakened intermolecular force (J.S., Cini, 1988) (V.I., 1984). That is to say, when a liquid expands, the intermolecular force is weakened, so the susceptibility is believed to be strengthened. Therefore, it is possible to determine the degree of the intensity of the interaction between water molecules and Na⁺·Cl⁻ ions by measurement of the diamagnetic susceptibility of NaCl solution. Then, we tried to investigate the difference in terms of NaCl concentration for the actual interaction force between water molecules and ions by comparing the measured value of susceptibility with the calculated value by the Pascal additivity law (T. Kurozawa, 1996), in the case of no intermolecular interaction. The calculation formula of susceptibility for NaCl concentration based on the Pascal additivity law is given by

\[ \chi = a \chi_w + b \chi_i + c \chi_c. \]  

where \( \chi, \chi_w, \chi_i, \) and \( \chi_c \) are the susceptibilities of NaCl solution, water, hydrated Na⁺·Cl⁻ ions and NaCl crystal, respectively. The measured values -6.67\( \times 10^{-7} \) emu/g and -4.85\( \times 10^{-7} \) emu/g were used in \( \chi_w \) and \( \chi_c \), respectively, and the calculated value -5.30\( \times 10^{-7} \) emu/g was used in \( \chi_i \) on the assumption that the susceptibility is proportional to the specific gravity of Na⁺ ion and Cl⁻ ion in these susceptibilities (Landolt, 1981). The coefficients \( a, b \) and \( c \) show the mass ratio of each component to NaCl solution, and \( a + b + c = 1 \). Table 1 shows each coefficient value for the concentrations of the NaCl solution. The value calculated by eq. (1) is shown by the dashed line in Fig. 4. Though the calculated value gradually decreases with increase in the NaCl concentration, the rapid reduction over the saturated concentration (26.45% at 25°C) is not observed. The latter is considered to be due to the small value of the coefficient \( c \) even if the NaCl concentration exceeded the saturated value. Naturally there is a gap between the measured value and the value calculated by eq. (1) in this figure. Because the change of the interaction force between water molecules and ions by differing NaCl concentration was not assumed in eq. (1). In fact, most of the interaction force between water molecules and ions is Coulomb force. According to our calculation, the Coulomb force was about 6.2 times (for Na⁺·H₂O) and about 2.4 times (for Cl⁻·H₂O) larger than the force between the two water molecules (H. Udaira, 1998) (J.N., 1992). Therefore, the interaction force between water molecules and hydrated Na⁺·Cl⁻ ions was stronger than that between both water molecules, and the binding condition was believed to be strengthened with increasing NaCl concentration. Thus, it is necessary to consider the change of the interaction force for each component in the solution in eq. (1) in order to explain the behavior of the measured value under differing NaCl concentration.

Table 1 Coefficient value of \( a, b, c \) and \( a_0, a_1 \) of NaCl concentration.

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<th>3.50</th>
<th>4.00</th>
<th>5.00</th>
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<td>0.0500</td>
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<td>0.08406</td>
<td>0.08141</td>
<td>0.07875</td>
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<tr>
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<td>0</td>
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<td>0</td>
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<td>0</td>
<td>0</td>
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<td>0</td>
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<td>0</td>
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<tr>
<td>( a_1 )</td>
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coefficient \(a\) of water in eq. (1) is obtained, it is necessary to distinguish between the water molecule which constructs the regular structure and that which interacts with a neighboring ion, because the susceptibility of water changes even in the same water molecule by the interaction with a neighboring ion (J.S., 1980) (R. Ciri, 1968) (V.J., 1984). Fig. 5 shows the well-known hydration model\(^{19}\) for the interaction between an ion and solvent in NaCl solution. According to this model, three regions were proposed: the circular region where water molecules are oriented toward the neighboring ions (Primary Hydration Shell, \(a\) in Fig. 5) the region with pure water structure (Water Bulk, \(c\) in Fig. 5) and the region (V.I., 1984) (H. Uedaira, 1998) (J.N., 1992) (H.S., 1957) where the water structure (hydrogen bond, \(b\) in Fig. 5) is destroyed between the Primary Hydration Shell and Water Bulk (Secondary Hydration Shell). To distinguish the water molecules, that in the Primary Hydration Shell was proposed as a water molecule which interacts with a neighboring ion, and those other than in the Primary Hydration Shell were assumed to be water molecules with a pure structure. Here, the influence of water molecule in the Secondary Hydration Shell was neglected as it was a weak influence.

From the fact that the vapor pressure \(P^v\) of the solvent (pure water) in NaCl solution decreases with increase in the concentration (Handbook, 2000), the relationship between the ratio of susceptibility and ratio of vapor pressure based on the vapor pressure \(P^v\) of pure water was obtained. The details will be given in the Appendix. Using the relationship, the calculation formula of susceptibility in a concentration up to the saturated condition was corrected by

\[
\chi = a_0 \chi_w + a_1 \chi_\text{w} \left( \frac{P^v}{P^v_\text{w}} \right)^{\frac{1}{3}} + b \chi_i ,
\]

where the coefficients \(a_0, a_1\) denote the mass ratio of water molecule which has the structure of pure water and that of a water molecule which interacts with ion, respectively, where \(a_0 + a_1 = a\) and \(a + b = 1 (c = 0)\). The result (H. Uedaira, 1998) of the X-ray diffraction method was used to obtain the coefficient \(a_1\). Namely, 8 and 6 water molecules were assumed to be orientated in Primary Hydration Shells of a Na\(^+\) and a Cl\(^-\) ion, respectively. Each coefficient value for the concentration of the NaCl solution is shown in Table 1. The value calculated by eq. (2) is shown by the dot-dash line in Fig. 4. The gap between the calculated value and our measured value was small. On the other hand, the calculated value clearly disagreed with the value of Hochart. Therefore, our measured value was recognized to be more reliable in explaining the behavior of susceptibility depending on NaCl concentration.

In the meantime, the interaction force in both ions is considered to strongly influence the rapid reduction of the susceptibility of NaCl 26.6%: When the NaCl concentration is over 26%, the average radius of an ion occupied in the solution can be smaller than 0.33 nm as obtained by the calculation. Then, the hydration shell of the Na\(^+\) and Cl\(^-\) ion are destroyed in either case (V.I., 1984). The Coulomb force inversely proportioned to the square of the distance between Na\(^+\) ion and Cl\(^-\) ion increased rapidly. The rapid increase of the Coulomb force increased the intermolecular force in NaCl solution (J.S, 1980) (R. Ciri, 1968) (V.I., 1984), therefore, the diamagnetic susceptibility rapidly decreased.

Finally, the concentration dependence of the gas-liquid interface deformation of the NaCl solution is discussed. The decreased level of diamagnetic susceptibility \(\chi\) by increasing the NaCl concentration within concentration of saturated solution was 9.9%, and it was smaller than a 16.8% increase of the density (Handbook, 2000) \(\rho\). It was found that the concentration dependence of the susceptibility could not be disregarded for deformed variation of the gas-liquid interface of the NaCl solution, because this deformation is proportional to \(\chi/\rho\).

5. Conclusions

The ampoule cell with good tightening was produced, and the diamagnetic susceptibility of NaCl aqueous solution over a wide concentration range was measured by using SQUID magnetometer. The measurement was performed
within a magnetic field of 6 T at 25±0.05°C for seven types of NaCl solutions of varying concentrations. Magnetization curves of water and NaCl solutions were linear within the maximum field of 6 T. The susceptibility was obtained from the gradient of the straight lines. As a result, the susceptibility of NaCl solution was less than that of water. The susceptibility of NaCl solution in the concentration from 0.262% (concentration of saturated solution) gently decreased as the concentration increased. Also, there was a rapid reduction of the measured value as the NaCl concentration was increased over the saturated concentration. Using the relationship between the susceptibility and the vapor pressure, the calculated value of the susceptibility was compared with our measured value. As a result, the measurement value was almost in accordance with the calculated value. It can be concluded that the magnetic susceptibility of the NaCl solution shows a decrease of approximately 10% within concentration of saturated solution, and that the susceptibility is one of the effective cause for the concentration dependence on the gas-liquid interface deformation of the NaCl solution.

References


Appendix

The ion concentration dependence for the microscopic change of chemical potential \( \mu \) on diamagnetic aqueous solution with the magnetic homogeneity (hereafter diamagnetic solution) in a uniform magnetic field is considered. The solvent in the solution is pure water and the solutes are ions. We assumed that the solution is electrically neutral, and there is no magnetic anisotropy. The ion concentration in the equation of \( \mu \) is expressed using the mole fraction \( X \) (the ratio of solvent mole number to the sum of solute and solvent mole number) of the solvent. Since \( X \) is related to pressure \( P \) and susceptibility \( \chi \), the \( \mu \) considers the change of the ion concentration in isothermal and identical magnetic field is given from Gibbs-Duhem relation(LEB, 1957).

\[
\mu = \frac{N}{V} dP(X) - \frac{B^2}{\mu_0 N} d\chi(X), \quad (A \cdot 1)
\]

where \( \mu \) is chemical potential, \( T \) is absolute temperature, \( B \) is magnetic field, \( V \) is unit volume, \( N \) is particle number in the unit volume and \( \mu_0 \) is permeability in the vacuum. For obtaining this equation, it was assumed that the susceptibility \( \chi \) of a diamagnetic solution did not depend on the magnetic field (see Fig. 3). The difference of chemical potential \( \mu_s \) (the subscript \( s \) stands for the solvent) of the solvent by the change from the mole fraction \( X=1 \) (pure water) to any concentration \( X \) in the solution up to a concentration of saturated solution was obtained from the positive integral of eq. (A \cdot 1) as

\[
\mu_s = \mu_s \left( P(X), \chi(X) \right)_{T,B} - \mu_s \left( P(1), \chi(1) \right)_{T,B} = \int_{X=1}^{X} \frac{N}{V} dP(X) - \int_{X=1}^{X} \frac{B^2}{\mu_0 N} d\chi(X), \quad (A \cdot 2)
\]

where \( P, \chi \) and \( X \) are vapor pressure, susceptibility and mole fraction of the solvent, respectively. The relationship between the susceptibility of the solvent and
the vapor pressure by the change of ionic concentration can be obtained using eq. (A · 2). It is assumed that the solution is nonvolatile and the vapor phase consists only of solvent molecules. Vapor pressure $P^v$ can be the internal pressure of the vapor phase of the solvent on this assumption. Then, the chemical potential of vapor phase of the solvent, $\mu^v_0$ (the subscript $v$ stands for the vapor), equilibrated with the solution was obtained from eq. (A · 2) as

$$\mu^v_0 \left( P^v(x), x(x) \right)_{T,B} - \mu^v_0 \left( P^v(1), x(1) \right)_{T,B} = \int_{x=1}^{X} kT \cdot \frac{dP^v(x)}{P^v(x)} - \frac{B^2}{\mu^2_0 N} dx(x), \quad (A \cdot 3)$$

where $k$ is the Boltzmann constant. When the chemical potential of vapor phase of pure water was indicated $\mu^v_w$ (the subscript $w$ stands for the water), since it was $\mu^v_w \left( P^v(1), x(1) \right)_{T,B} = \mu^v_w \left( P^v(1), x(1) \right)_{T,B}$, the $\mu^v_0$ in the second term on the left-hand side was converted into $\mu^v_w$ in order to obtain eq. (A · 3). Also, it was assumed that the vapor phase of the solvent behaved as an ideal gas in which there was seldom any interaction among water molecules at room temperature and at atmospheric pressure, $V/N = kT / P^v$ from the ideal gas state equation of $P^v = NkT$ was used in the first term on right-hand side of eq. (A · 3). When the vapor pressure of the solvent is equal to atmospheric pressure $P_e$ (or external pressure), namely $P^v = P_e$, the chemical potential of vapor phase of the solvent can be $\mu^v_0 \left( P_e(x), x(x) \right)_{T,B} - \mu^v_0 \left( P_e(1), x(1) \right)_{T,B}$. (D. Kondapui, 1998) Therefore, the relationship between the susceptibility of the solvent and vapor pressure was obtained from eq. (A · 3) as

$$\frac{B^2}{\mu^2_0 N} dx = kT \cdot \frac{dP^v}{P^v}. \quad (A \cdot 4)$$

To obtain the relationship between susceptibility $\chi$ and vapor pressure $P^v$ in isothermal and identical magnetic fields, eq. (A · 4) has to be changed to an equation which does not depend on magnetic field and temperature. Thus, it is necessary to note the relationship between temperature $T$ and magnetic field $B$. Then, the relationship between $B$ and $T$ in the isothermal and identical magnetic fields was obtained using Curie's Law (W. Greiner, 1995) for the diamagnetic molecule. As the result, the relationship became $kT = \chi B^2 / 3 \mu_0 N$ in terms of $\chi$. As this relation was substituted in eq. (A · 4), the relationship in which the vapor pressure $P^v$ and the susceptibility $\chi$ does not depend on the magnetic field and the temperature was given by

$$\frac{d\chi}{\chi} = \frac{1}{3} \frac{dP^v}{P^v}. \quad (A \cdot 5)$$

Both sides in eq. (A · 5) can be integrated positively from the mole fraction $X = 1$ (pure water) to any concentration $X$ in the solution up to a concentration of saturated solution. Also, when the susceptibility and vapor pressure of pure water are taken as $\chi^v(1) = \chi^v_w$ and $P^v(1) = P^v_w$, and the susceptibility and vapor pressure in some mole fraction $X$ are taken as $\chi(X) = \chi$ and $P(X) = P$, susceptibility of the solvent from the change of vapor pressure is obtained by

$$\left( \frac{\chi}{\chi_w} \right) = \left( \frac{P^v}{P^v_w} \right)^{\frac{1}{3}}. \quad (A \cdot 6)$$

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