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Stereoelectronic Structure of 3-(Trichlorogermyl)propionic Acid by the Results of *ab initio* Calculations

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Abstract—Two structures of the 3-(trichlorogermyl)propionic acid molecule and its dimer were calculated by the RHF/6-31G(d) method with the full geometry optimization. The structure with pentacoordinated germanium atom is by 4.71 kcal mol^{-1} more favorable than that with tetracoordinated germanium. The strength of coordination bond in the first structure increases with the absolute values of charge on the Ge and O coordination centers. The relatively small values of these charges result in a weak coordination bond. In the first structure, this bond is weaker than in the dimer, since the Ge···O distance in it (3.016 Å) is larger than in the latter (2.898 Å). This bond is formed due to the rapproachment of the Ge and O coordination centers at their electrostatic interaction. This provides the transfer of electron density from the atoms of the donor fragment of the molecule to the atoms of the germanium coordination polyhedron. The coordination centers serve as the conductors for the electron density transfer.

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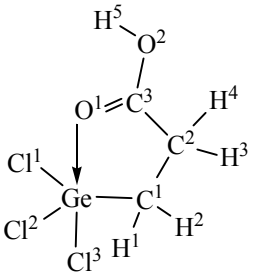
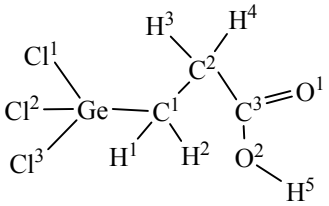
The X-ray data [1, 2] indicate that the Ge←O coordination interaction in the molecule of 3-(trichlorogermyl)propionic acid (**I**) is very weak. An argument in favor of such interaction is the conformation of the fragment Ge–CH_2–CH_2–COOH, in which the Ge and O atoms are at a distance 3.228 Å, less than the sum of their van der Waals radii, despite the fact that in this fragment there is a possibility of rotation around three single bonds. It is suggested that this coordination interaction is weak due to the presence of electron–acceptor hydroxy group and hydrogen bonds that combine the molecules of **I** into dimers [1]. The coordination polyhedron of the germanium atom has tetrahedral structure [1, 2]. The ^{35}Cl nuclear quadrupole resonance (NQR) spectrum of this compound measured at 77 K consists of three lines of equal intensity close by frequency (23.550, 22.695, and 22.444 MHz) [3, 4] that does not correspond to the trigonal-bipyramidal structure of this compound although is consistent with the pentacoordination of the Ge atom [3, 4].

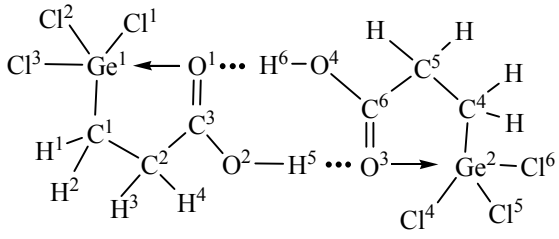
The aim of this work was to study the nature of coordination bond in this molecule, changes in its stereoelectronic structure at the change in the distance between coordination centers, and the effect of dimerization on this coordination bond. For this purpose, we performed complete quantum-chemical calculations of



I



II



III

Table 1. Bond lengths (*d*), bond (ω) and torsion (τ) angles in molecule **I**, calculated by the RHF/6-31G(d), and XRD data [1, 2]

Bond	*d*, Å	d_{PCA}, Å	Angle	ω, deg	ω_{PCA}, deg	Angle	τ, deg
$Ge–Cl^1$	2.133	2.121	Cl^1GeCl^3	104.96	105.72	$Cl^1GeC^1C^2$	–108.37
$Ge–Cl^2$	2.146	2.123	Cl^2GeCl^3	104.88	104.35	$Cl^2GeC^1C^2$	19.38
$Ge–Cl^3$	2.159	2.134	Cl^1GeCl^2	109.33	106.58	$Cl^3GeC^1C^2$	135.08
$Ge–C^1$	1.938	1.929	Cl^3GeC^1	107.32	108.70	$GeC^1C^2C^3$	59.69
$C^1–C^2$	1.527	1.528	Cl^1GeC^1	115.51	117.00	$C^1C^2C^3O^1$	–11.87
$C^2–C^3$	1.505	1.500	Cl^2GeC^1	113.86	113.50	$C^1C^2C^3O^2$	168.95
$C^3–O^1$	1.190	1.203	GeC^1Cl^2	115.52	116.20	$C^2C^3O^2H^5$	177.94
$C^3–O^2$	1.321	1.309	$C^1C^2C^3$	111.87	113.10	$O^1C^3O^2H^5$	–1.24
$Ge^1\cdots O^1$	3.015	3.228	$C^2C^3O^1$	123.90	123.10		
			$C^2C^3O^2$	112.87	113.10		
			$O^1C^3O^2$	123.23	123.70		

structures **I–III** by the RHF/6-31G(d) method with the program GAUSSIAN 03W [5] with the full optimization of their geometry, and of the structure **I** at various $Ge\cdots O^1$ distances. The origin of the coor-dinate system was chosen at the chlorine atom whose ^{35}Cl NQR parameters should be identified. The *Z* axis of the system coincides with the respective Ge–Cl bond.

The ^{35}Cl NQR frequencies (ν) and asymmetry parameters (η) of the electric field gradient (EFG) at the ^{35}Cl nuclei were calculated from the populations of the less diffuse 3*p*-components of valence *p* orbitals of chlorine atoms [6–9] by Eqs. (1) and (2), respectively [10].

$$\nu = (e^2Qq_{at}/2h[-N_z + (N_x + N_y)/2](1 + \eta^2/3)^{1/2}, \quad (1)$$

$$\eta = |3(N_x - N_y)/(2N_z - N_x - N_y)|. \quad (2)$$

Here e^2Qq_{at} is the constant of nuclear quadrupole interaction; *h* is the Planck's constant; N_x, N_y, and N_z are the populations of 3*p*-components of valence p_x, p_y, and p_z orbitals, respectively, of the tracer chlorine atom. The value of $e^2Qq_{at}/2h$ was found from the experimental NQR frequency of Cl_2 at 77 K, and the populations of 3*p*-components of valence *p* orbitals of the Cl atoms of this molecule were obtained by the calculation with an appropriate method [6–9].

The X-ray data and ^{35}Cl NQR spectra were obtained for the crystalline state of 3-(trichlorogermyl) propionic acid, in which the molecules not always are in the energetically most favorable form [11, 12]. So it is necessary to explore stereoelectronic structure of this acid in the gaseous state, and to compare it with existing X-ray diffraction data and the ^{35}Cl NQR spectrum.

According to the calculations, the energetically most favorable is the structure **I**. Its total energy with accounting for the zero point energy is by 4.71 kcal mol^{-1} lower than the energy of structure **II**. In addition, the latter structure corresponds to the saddle point of the second order on the potential energy surface (it has two imaginary frequencies of stretching vibrations). Judging from the bond angle values of the germanium atom, we conclude that the coordination polyhedron in structure **I** is closer to the tetrahedron than to the trigonal bipyramid. Some of the calculated geometric characteristics of the structure **I** (Table 1) differ from the X-ray data [1, 2]. In particular, the calculated Ge–Cl and $C^3–O^2$ bond lengths are much longer than the experimental ones, while the $Ge\cdots O^1$ distance, on the contrary, is shorter. Consequently, the geometry of molecule **I** in gaseous and crystalline states is somewhat different. At the same time, in the crystalline state molecule **I** is energetically less favorable than in the gas. The electron distribution in these states should also be different. This is confirmed by ^{35}Cl NQR frequencies and asymmetry parameters of electric field gradient (EFG) on the ^{35}Cl nuclei of molecule **I** estimated with Eqs. (1) and (2), respectively, using the

Table 2. Populations of valence *p* orbitals of chlorine atoms (Σ*Np*) in molecules **I** and **II** calculated with full optimization of geometry using RHF/6-31G(d) method, the populations of 3*p*- and 4*p*-components (*N*) of these orbitals, and calculated from the populations of 3*p*-components NQR ^{35}Cl frequencies (ν_{calc}) and asymmetry parameters (η_{calc}) of electric field gradient EFG at the ^{35}Cl nuclei

Molecule	Atom	Orbital	N_x, *e*	N_y, *e*	N_z, *e*	ν_{calc}, MHz	η_{calc}, %
I	Cl^1	*N*3*p*	1.287	1.271	1.022	23.960	9.34
		*N*4*p*	0.641	0.650	0.382		
		Σ*Np*	1.928	1.921	1.404		
	Cl^2	*N*3*p*	1.271	1.279	1.031	22.724	4.92
		*N*4*p*	0.655	0.650	0.396		
		Σ*Np*	1.926	1.929	1.427		
	Cl^3	*N*3*p*	1.272	1.271	1.039	21.644	0.64
		*N*4*p*	0.656	0.656	0.394		
		Σ*Np*	1.928	1.927	1.433		
II[a]	Cl^1	*N*3*p*	1.273	1.275	1.032	22.529	1.24
		*N*4*p*	0.650	0.650	0.384		
		Σ*Np*	1.923	1.925	1.416		

[a] The data for only one chlorine atom, since the length of the three bonds Ge–Cl (2.141 Å) and angles ClGeCl between them (106.8°) are almost identical and, therefore, the electron distribution on all three chlorine atoms in structure **II** is virtually the same.

populations of 3*p*-components of valence *p* orbitals of chlorine atoms (Table 2) derived by quantum chemical calculations of this molecule. Since the bond length of different Ge–Cl bonds obtained by calculation of the molecule **I** with full optimization of its geometry differ markedly (Table 1), for a correct estimation of the populations we performed these calculations three times with the origin of the coordinate system located at each chlorine atom. The NQR frequency calculated in this way (Table 2) for the Cl^1 atom is only slightly higher than the highest frequency of the experimental line, the frequency of Cl^2 atom is close to the frequency of the medium line, and the frequency of Cl^3 atom is markedly lower than that of the lowest frequency of experimental lines. The electron distribution on the Cl^3 atom has nearly axial symmetry, and the NQR frequency of this atom is typical for the axial chlorine atoms in the molecules with the pentacoordinated germanium atom [3, 4, 13]. The NQR frequency calculated for the Cl^1 atom may relate to both the chlorine atoms associated with tetracoordinated Ge atom, and to the equatorial chlorine atoms in the molecules with pentacoordinated germanium atom. The NQR frequency of the Cl^2 atom in the structure **I** is close to the NQR frequency in the structure **II** with tetracoordinated germanium atom (Table 2).

The ^{35}Cl NQR frequencies (Table 3) evaluated from the populations of 3*p*-components of valence *p* orbitals of chlorine atoms in the structure **I** differ significantly from the experimental NQR data (see above). The calculations were performed using X-ray data (Table 1) [1, 2], with optimization of other geometric characteristics of the molecule (dihedral angles and some bond angles and bond lengths). Since the experimental length of the equatorial Ge–Cl^1 and Ge–Cl^2 bonds are almost identical and the corresponding bond angles of the germanium formed by these bonds are close to each other (Table 1), the electron distribution on these chlorine atoms should be almost identical. Therefore, the calculation of the NQR parameters was performed for only one equatorial chlorine atom. The calculated NQR frequencies are characteristic of the axial and equatorial chlorine atoms, respectively, in the trigonal–bipyramidal structure of organyltrichlorogermanes [3, 4, 13]. The difference between the calculated and experimental NQR data is due not only to

Table 3. The populations of 3*p*-components of valence *p* orbitals of chlorine atoms (*N*) in molecule **I**, calculated by RHF/6-31G(d) method at different Ge···O distances, and calculated from these populations NQR frequencies (ν_{calc}) and asymmetry parameters (η_{calc}) of electric field gradient at the ^{35}Cl nuclei

Atom	Parameter	*d*(Ge···O), Å						
		2.2	2.4	2.6	2.8	3.0	3.228[a]	3.4
Cl^1	$N3p_x$, *e*	1.296	1.296	1.294	1.291	1.287	1.284	1.281
	$N3p_y$, *e*	1.261	1.265	1.267	1.270	1.271	1.272	1.274
	$N3p_z$, *e*	1.022	1.019	1.019	1.020	1.022	1.028	1.026
	ν_{calc}, MHz	24.044	24.472	24.441	24.310	23.960	23.293	23.420
	η_{calc}, %	20.47	17.78	15.49	12.09	9.34	7.2	4.18
Cl^2	$N3p_x$, *e*	1.262	1.266	1.268	1.270	1.271	–	1.272
	$N3p_y$, *e*	1.291	1.288	1.285	1.282	1.279	–	1.275
	$N3p_z$, *e*	1.027	1.026	1.027	1.029	1.03	–	1.033
	ν_{calc}, MHz	23.344	23.433	23.267	23.014	22.817	–	22.390
	η_{calc}, %	17.43	13.15	10.22	7.29	4.90	–	1.87
Cl^3	$N3p_x$, *e*	1.264	1.267	1.269	1.271	1.272	1.273	1.273
	$N3p_y$, *e*	1.260	1.264	1.267	1.269	1.271	1.272	1.273
	$N3p_z$, *e*	1.051	1.048	1.045	1.042	1.039	1.042	1.035
	ν_{calc}, MHz	19.645	20.249	20.760	21.226	21.644	21.458	22.156
	η_{calc}, %	2.84	2.07	1.34	1.32	0.64	0.65	0.0

[a] Calculations are performed using XRD data [1, 2].

the partial use of X-ray data for the quantum-chemical calculations of molecules, but, apparently, due to the fact that the X-ray data and the NQR spectra were obtained at significantly different temperatures.

Previously it has been shown [3, 4] that one of the conditions for the pentacoordination of the atom of an element of IVA group is a significant positive charge on the atom and the negative charge on the heteroatom of organic substituent at this atom. The quantum-chemical calculation of molecule **I** with full optimization of its geometry and the Mulliken analysis of electron distribution showed that this condition is satisfied (Table 4): the germanium atom has a significant positive charge (0.707 *e*), and the carbonyl oxygen atom bears a negative charge (–0.555 *e*). However, these charges are much smaller than the charges on the atoms in the molecules of 3-trichlorogermylpropanoic amide (0.759 and –0.616 *e*, respectively [14]) and 2-methyl-3-(trichlorogermyl)propanoic *N,N*-dimethylamide (0.779 and –0.666 *e*, respectively [15]), calculated by the RHF/6-31G(d) method with complete optimization of molecular geometry. The increase in charges on the Ge and O atoms in the two amide molecules compared with molecule **I** is accompanied by a decrease in the calculated length of the coordination Ge···O bond: 3.015 in **I** (Table 1), 2.607 [14] and 2.321 Å [15] in the amides, respectively. The length of coordination bond in these molecules found by X-ray analysis vary similarly: 3.228, 2.166, and 2.123 Å, respectively [1–3]. Consequently, the strength of coordination bond increases with the increase in the absolute value of the charges on the Ge and O coordination centers. The relatively small magnitudes of these charges in molecule **I** explain why the coordination bond is weak. The difference in the values of the charges on the Ge and O atoms in these three molecules is due, above all, to the difference in substituents at the carbonyl carbon atom.

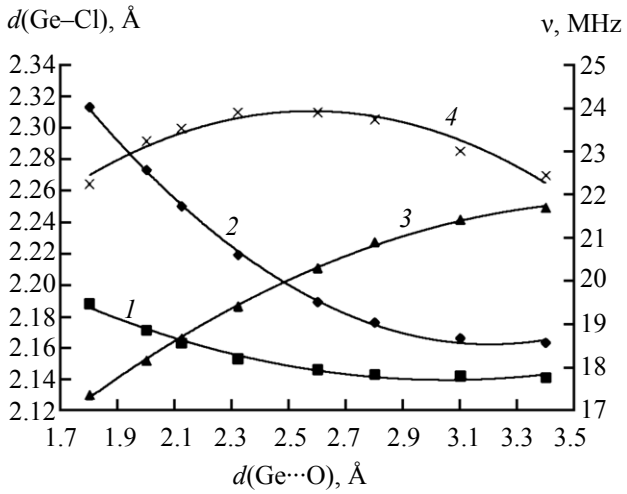
It is believed [1] that the weakening of the GeO coordination bond in molecule **I** is caused, in particular, by the hydrogen bonding that combines the

Table 4. Atomic charges (*q*) in molecule **I**, calculated by the RHF/6-31G(d) method at different Ge···O distances (*d*)

Atom	*q*, *e* at *d*(Ge···O), Å						
	2.2	2.4	2.6	2.8	3.0	3.015[a]	3.4
Cl^1	–0.227	–0.216	–0.209	–0.204	–0.200	–0.200	–0.195
Cl^2	–0.246	–0.241	–0.238	–0.236	–0.233	–0.232	–0.224
Cl^3	–0.305	–0.284	–0.267	–0.251	–0.234	–0.233	–0.214
Ge	0.775	0.762	0.749	0.731	0.708	0.707	0.673
C^1	–0.565	–0.559	–0.555	–0.552	–0.549	–0.549	–0.550
C^2	–0.428	–0.430	–0.431	–0.430	–0.427	–0.427	–0.422
C^3	0.813	0.803	0.794	0.785	0.776	0.776	0.765
O^1	–0.591	–0.582	–0.572	–0.563	–0.556	–0.555	–0.550
O^2	–0.667	–0.677	–0.684	–0.689	–0.691	–0.691	–0.693
H^1	0.241	0.239	0.237	0.235	0.234	0.233	0.231
H^2	0.230	0.227	0.227	0.229	0.233	0.233	0.244
H^3	0.237	0.233	0.231	0.229	0.229	0.229	0.229
H^4	0.243	0.240	0.238	0.237	0.236	0.236	0.234
H^5	0.490	0.484	0.480	0.477	0.476	0.476	0.474

[a] Calculation with full geometry optimization.

molecules into dimer **III**. However, judging from the values of the Ge···O distances derived from calculations of the dimer this distance (2.898 Å) is shorter than that in the individual molecule (Table 1) and, therefore, makes the dimer coordination bond somewhat stronger. Judging from the total energy of the dimer **III** (taking into account its zero point energy, it is by 14.44 kcal mol^{-1} lower than the sum of total energies of the two individual molecules), it is energetically more favorable than structure **I**. The formation of hydrogen bonds (O···H distance in the dimer 1.834 Å) has almost no effect on the Ge–Cl bond lengths (2.138, 2.145 and 2.159, Å) and calculated ^{35}Cl NQR frequencies (23.723, 22.910, and 21.644 MHz for atoms Cl^1, Cl^2 and Cl^3, respectively). Therewith some increase occurs in the bond lengths O–H (0.966 Å) and C=O (1.205 Å) compared with structure **I** and insignificant change of almost all valence and dihedral angles.



Dependence on the Ge···O distance of calculated (*1*) axial and (*2*) the average equatorial bond lengths of P–Cl bonds in molecule **I**, as well as the ^{35}Cl NQR frequencies of (*3*) axial and (*4*) equatorial chlorine atoms.

The Ge←O donor-acceptor interaction in molecule **I** should diminish the negative charge on the O atom and the positive charge of the atom Ge. However, in structure **I** with weak Ge←O coordination interaction the charges on these atoms increase (Table 4) compared with those in structure **II** in which the coordination interaction is absent and the charge on the atoms are: O^1 –0.549 *e*, Ge 0.656 *e*.

While shortening the Ge···O distance from 3.4 to 2.2 Å and optimizing the remaining geometric characteristics of molecule **I** the calculated length of the equatorial Ge–Cl bonds increases slightly, whereas the length of the axial Ge–Cl increases significantly. The relationship between the length of Ge···O and Ge–Cl bonds is not linear but exponential (see the figure), like that found for 2-methyl-3-(trichlorogermyl) propionic acid *N*,*N*-dimethylamide [15] and found experi-mentally for Ge–Cl and Ge···O bond lengths in the molecules containing Cl_3Ge–CCC(O)X fragment [13]. For both equatorial Ge–Cl bonds this dependence is the same. The figure shows average length of these two bonds. With decreasing Ge···O distance in the molecule **I** the calculated NQR frequencies of axial and equatorial chlorine atoms vary linearly (Table 3, the figure) like the NQR frequencies of the equatorial and axial chlorine atoms in 2-methyl-3-(trichlorogermyl)-propionic acid *N*,*N*-dimethylamide [15]. The variations of the NQR frequencies of the two equatorial chlorine atoms are identical. Therefore, the figure shows the dependence on the distance Ge···O only for the Cl^1 atom. The data show that the axial Ge–Cl bond and axial chlorine atom are much more sensitive to changes in the Ge···O distance than the equatorial ones.

We have previously shown [14, 15] that at the rapproachment of the coordination centers of Ge and O in organyltrichlorogermanium compounds the partial positive charge on the germanium atom and partial negative charge on the oxygen atom of the organic substituent increase, which cannot be regarded as a transfer of electron density in these molecules from the oxygen atom to the germanium atom. These atoms serve only as conductors of electron density from the electron–donor fragment of the molecule on the elec-tron–acceptor one. The same is observed in molecule **I**. With this rapproachment the negative charges on the chlorine atoms and C^1 also increase. Apparently, this increase is due to a shift of electron density from the germanium atom, as well as from the atoms C^3, O^2, and H^5, on which the electron density, conversely, decreases at mutual approaching of the coordination centers (Table 4). At this rapproachment the charge on the atom C^2 remains almost unchanged.

Thus, at the rapproachment of the coordination centers Ge and O as a result of their electrostatic interactions, in molecule **I** a weak Ge←O bond is formed, which transfers electron density from the atoms of the donor fragment of the molecule to the atoms of the germanium coordination polyhedron, in the same way as at the formation of complex compounds of IVA group element with organic ligands the electron density of atoms of the ligand is transferred to the electron acceptor atoms [16–19]. Coordination centers in those and other molecules serve as conductors for transfer of the electron density.

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