

Spin Relaxation and Magnetic Properties of Benzo-1,2,3-tritholium Radical Cations

V. I. Krinichnyi¹, R. Herrmann², E. Fanghaenel²,
W. Moerke², and K. Lueders³

¹Institute for Chemical Physics, Chernogolovka, Russian Federation

²Martin-Luther-Universität Halle-Wittenberg, Merseburg, Germany

³Hochschule für Technik, Wirtschaft und Kultur, Leipzig, Germany

Received December 25, 1995; revised December 2, 1996

Abstract. Magnetic and dynamics properties of paramagnetic centers in various tritholium cations were characterized by static magnetic susceptibility measurements and EPR spectroscopy. Magnetically correlated units consisting of at least pairs of spins with ferromagnetic exchange coupling were found in some trithioles. The rates of intramolecular spin diffusion and intermolecular spin hopping were estimated separately. The rates and the anisotropy ($\nu_{\parallel}/\nu_{\perp} = 20\text{--}300$) of spin dynamics were shown to depend on the molecular structure.

1. Introduction

Sulfur in organic compounds enables a great number of cyclic systems. One example are the used 1,2,3-Benzotrithioles with the general structure $\overline{\text{RCSSSCR}}$ [1, 2] and stability that differs essentially from that of other cyclic trisulfides, which are known to be highly unstable [3]. These trithioles form radical cations with strong oxidizing agent. These radical cations have some interesting and unusual physical properties, which could be important for the creation of new organic magnetic and conducting materials. Therefore the study of various properties of such substances may be useful for the synthesis of compounds with controlled magnetic and electron transport properties. $\text{F}_3\overline{\text{CCSSSCCF}_3^+}\text{AsF}_6^-$, reported by Cameron *et al.* [1, 2], is a simple tritholium salt with both spin and positive charge localized on the trisulfuric group. An unpaired electron interacts with zero-spin sulfur nucleus resulting in a shift of its g -factor to 2.014 [1].

We present in this communication the results of investigations of magnetic, spin-relaxation and dynamic properties of the new benzo-1,2,3-tritholium radicals (Fig. 1).

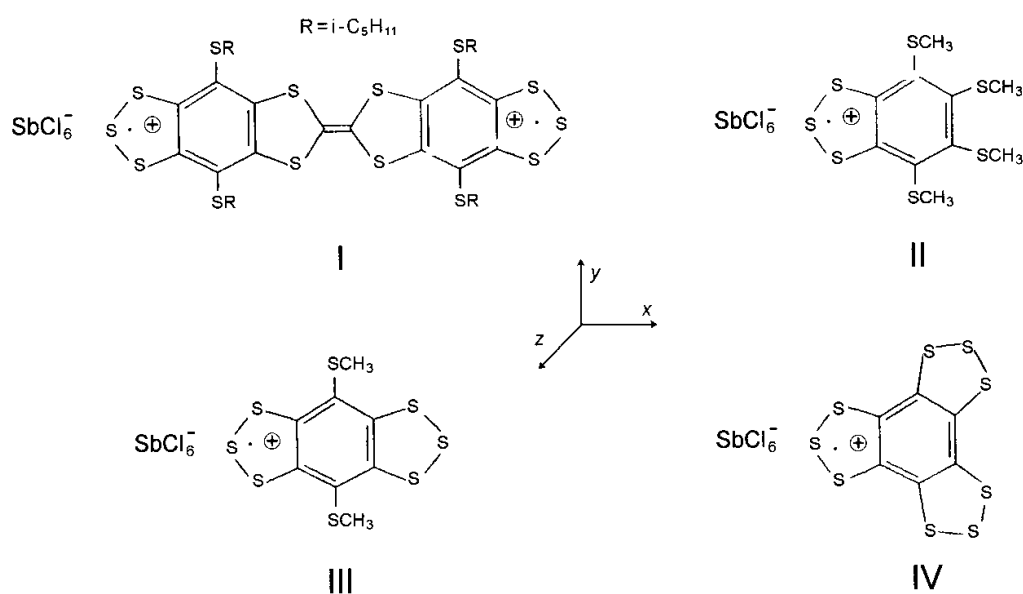


Fig. 1. Benzo-1,2,3-tritholium radicals.

2. Experimental

The powder-like tritholium samples I–IV were synthesized as described in detail in [4]. They were placed into EPR probe quartz tubes filled with nitrogen as sample protector and heat exchange gas. EPR measurements were performed in the 93–300 K temperature range with a commercial X-band ($\nu_e = 9.4$ GHz) ZWG ERS 200 spectrometer. At each temperature setting realized by an appropriate gas-flow variable temperature cryosystem, the signals of the sample and the lateral ruby standard were recorded simultaneously. The same standard was also used for setting the microwave field. Thus absolute EPR susceptibility, the positions of g -tensor components and the linewidth of the spectral components were exactly determined for each sample. The accuracy of these values is $2 \cdot 10^{-7}$ emu/mole, $2 \cdot 10^{-4}$ and $1 \cdot 10^{-2}$ mT, respectively.

The static (total) magnetic susceptibility of the samples was measured with an accuracy of $3 \cdot 10^{-7}$ emu/mole by using a LAKESHORE 7221 ac susceptometer in the 4–300 K temperature range at a maximum field strength of 1 T.

3. Results and Discussion

The static (total) magnetic susceptibility χ consists of the temperature dependent (Curie) contribution of orientation paramagnetism χ_{po} and the temperature independent contribution of Van Vleck polarization paramagnetism χ_{pp} arising due to molecular asymmetry and precession diamagnetism χ_d [5]:

$$\chi = \chi_{po} + \chi_{pp} + \chi_d = \frac{N \mu_0 \mu_B^2 p^2}{3k_B T} + \frac{2N_A}{3} \sum_i \frac{M_z^2}{\Delta E_i^{(0)}} - \frac{N_A \mu_0 e^2}{6m_e} \sum_j \langle r_i \rangle, \quad (1)$$

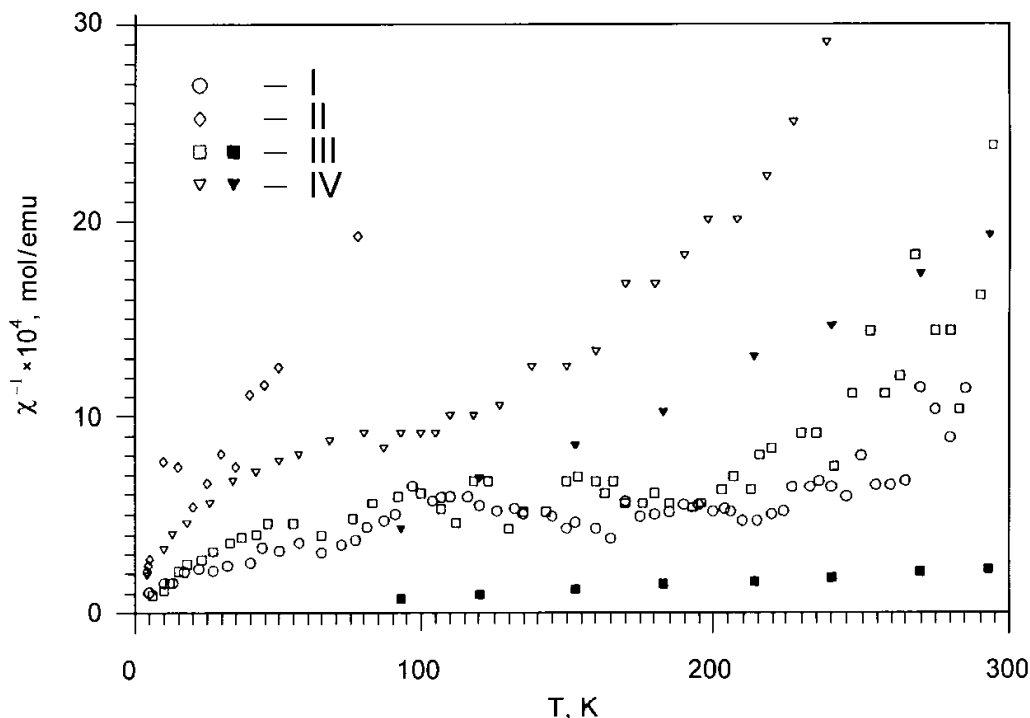


Fig. 2. The temperature dependences of reversed magnetic susceptibility of benzo-1,2,3-trithiolium salts determined by ac susceptometry (open points) and EPR (closed points).

where N is the volume spin concentration, μ_0 is the permeability for vacuum, p is the effective number of Bohr magnetons μ_B per molecule, k_B is the Boltzmann constant, T is the absolute temperature, N_A is the Avogadro number, M_z is the matrix element of the operator of the magnetic moment \mathbf{M}_z , $\Delta E_i^{(0)}$ is the energy distance between ground and excited states, e and m_e are the charge and mass of electron, respectively, and $\langle r_i \rangle$ is the averaged radius of the trajectory of an electron rotation.

Figure 2 shows the temperature dependences of χ for the samples I–IV determined using ac susceptometer and corrected for the diamagnetic contribution, and the same dependences for the samples III and IV obtained by X-band EPR spectroscopy. The slopes and characters of the functions determined by both methods display differences for some samples. This was probably caused by the paramagnetic saturation [5] in the external magnetic field of $B_0 \geq 330$ mT. On the other hand, the susceptibility evaluated from double integration of the EPR line can be influenced not only by the concentration of the spins but also by their temperature dependent relaxation and exchange rates. There may be an additional reason for altering the EPR susceptibility data and accordingly for above mentioned discrepancy. Evidently the $\chi(T)$ dependence determined by ac susceptometry for the sample II follows a simple Curie law $\chi(T) \sim T^{-1}$, whereas the other slopes are characterized by more complex functions.

Assuming that χ_{po} predominates in χ , the p value can be determined from the data shown in Fig. 2. Figure 3 presents these values versus temperature. The effective number of magnetons in sample II is practically temperature

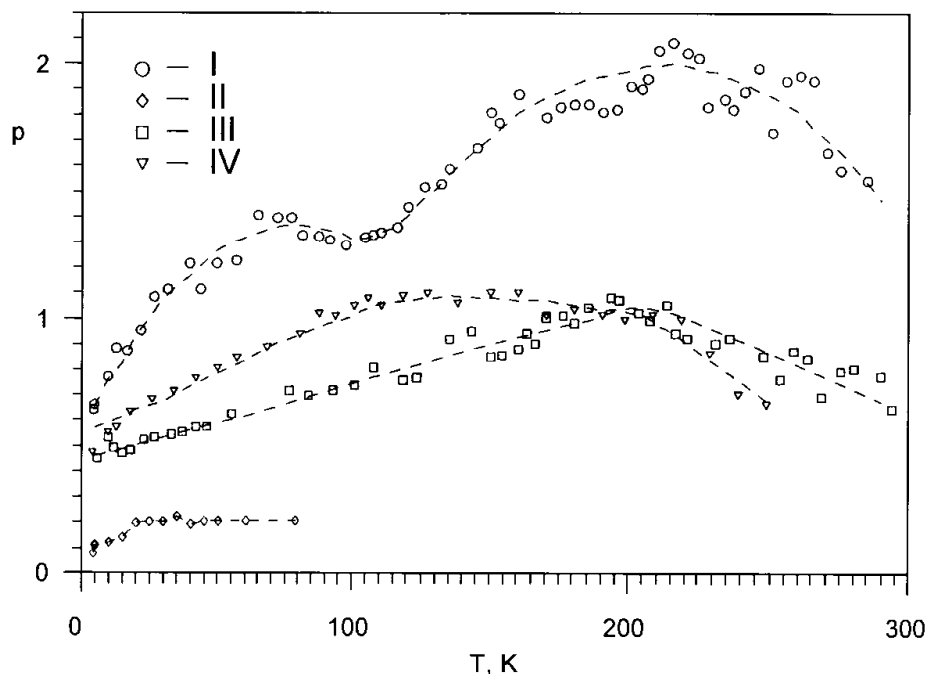


Fig. 3. The plots of effective number of Bohr magnetons μ_B per molecule of the benzo-1,2,3-trithiolium salts.

independent but in the other samples it follows the temperature function with extremes. The powders **III** and **IV** demonstrate the extremes of $p(T)$, for $\partial p(T)/\partial T = 0$ approximately at 200 and 140 K, respectively. The function for the sample **I** shows at least two extremes approximately at 70 and 210 K (see Fig. 3). The increase of the p value as temperature decreases in high-temperature range can probably be explained by the growth of the energy distance between the ground and nearest excited states ΔE_{01} , so that p increases from one limit $p = \sqrt{L(L+1) + 4S(S+1)}$ when $\Delta E_{01} \ll k_B T$ to another one $p = \langle g \rangle \sqrt{L(L+1) + 2\sqrt{L(L+1)S(S+1)} + S(S+1)}$ at $\Delta E_{01} \gg k_B T$ [5]. Here L and S are the orbital and spin moments, respectively, and $\langle g \rangle = (g_{xx} + g_{yy} + g_{zz})/3$ is the average g -factor. The decrease of magnetic susceptibility with temperature is typical for ferromagnetic materials at low temperatures [5, 6] and probably results from the growth of the spin exchange in the system. Assuming that stronger spin exchange exists in all samples compared with the sample **I**, it is possible to write for the low-temperature range [5]

$$p = \langle g \rangle J \frac{\exp(-x)}{1 + \exp(-x)}, \quad (2)$$

where J is the total moment of an electron, and $x = \Delta E_{01}/(k_B T)$. Note that the analogous behavior follows also from the $\chi(T)$ data for the $\overline{\text{F}_3\text{CCSSSCCF}_3^+\text{AsF}_6^-}$ salt in the low temperature range [2].

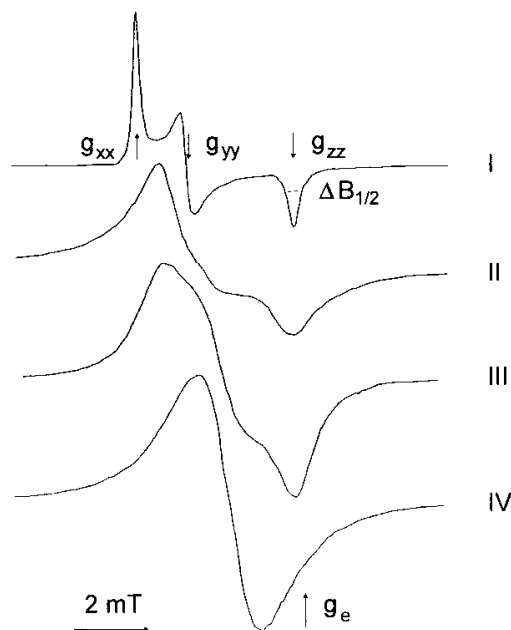


Fig. 4. X-band EPR absorption spectra of the benzo-1,2,3-trithiolium radicals registered at room temperature under nitrogen gas. The magnetic parameters measured and the position of the g -factor for free electron $g_e = 2.00232$ are shown.

Figure 4 shows the absorption X-band EPR spectra of the investigated samples. The line shapes of all EPR spectra are Lorentzian as shown by computer simulation. This behavior is typical for magnetic diluted system with chaotically localized paramagnetic centers and therefore the rate of spin diffusion ν_{diff} exceeds the linewidth at half-height $\Delta B_{1/2}$ [7]. The magnetic parameters of the compounds are summarized in Table 1.

Apparently sample I has a strongly anisotropic EPR spectrum (see Fig. 4) with $g_{xx} = 2.03230$, $g_{yy} = 2.0231$ and $g_{zz} = 2.00371$, and linewidths of components at half-height $\Delta B_{xx} = 0.19$, $\Delta B_{yy} = 0.55$ and $\Delta B_{zz} = 0.36$ mT. This spectrum is characteristic for polyoriented radicals with intramolecular coupling of the localized spin with sulfur heteroatoms possessing high spin-orbit constant λ_s and zero nuclear spin. As each molecule possesses two unpaired electrons with spin $S = 1/2$ [8], their dipole triplet state can be described by the Hamiltonian

$$\mathcal{H} = \mathbf{S} \mathbf{D} \mathbf{S} = D(S_z^2 - \frac{1}{3}S^2) + E(S_x^2 - S_y^2) , \quad (3)$$

where S_i are the canonic components of the spin tensor \mathbf{S} , and D and E are the independent constants. This interaction should split the components of the spectrum in magnetic field units by value [9]

$$\delta B = \frac{D}{\gamma_e \hbar} = \frac{3}{4} \left(\frac{\mu_0}{4\pi} \right) \gamma_e \hbar r^{-3} (1 - 3 \cos^2 \theta) , \quad (4)$$

Table 1. Parameters of the samples determined at 300 (120) K.

Parameter	Sample			
	I	II	III	IV
N (10^{23} m^{-3})	2.5	3.1	14	4.2
r_0 (nm)	0.74	0.77	0.61	0.53
R (nm)	0.85	0.42	0.40	0.40
χ_D (10^{-6} emu/mole)	-25	-9.7	-6.2	-8.3
$\langle g \rangle$	2.01970 (2.01795)	2.01694 ^a (2.01497) ^a	2.01442 ^a (2.01185) ^a	2.01510 (2.01433)
$\langle \Delta B_{1/2} \rangle^b$ (mT)	0.37 (0.32)	1.33 ^a (2.18) ^a	1.23 ^a (0.97) ^a	1.55 (1.12)
M_2 (mT ²)	0.083	0.092	0.83	0.38
ν_{\parallel} (10^8 s^{-1})	—	1.3	4.4	11.0
ν_{\perp} (10^6 s^{-1})	1.8	6.2	16	3.4

^a Determined by computer simulation of the spectrum.

^b Determined by using the equation $\langle \Delta B_{1/2} \rangle = (\Delta B_{1/2}^x + \Delta B_{1/2}^y + \Delta B_{1/2}^z)/3$.

where γ_e is the gyromagnetic ratio, $\hbar = h/2\pi$ is the Plank constant, r is the distance between the neighboring spins, and θ is the angle between the direction of the external magnetic field B_0 and molecular X axis. Using $r = 1.44 \text{ nm}$ [8] and averaging $(1 - 3\cos^2\theta)$ over sphere for D value averaging, $\delta B = 1.42 \text{ mT}$ is obtained. This value is really considerably higher than the halfwidth of all spectral components. Therefore, the transfer integral and hence the D constant are very small compared with the quantum energy $h\nu = \gamma_e \hbar B_0$ to consider cation **I** as a biradical. Accordingly this system can be considered as two separated and non-interacting radicals with $S = 1/2$.

In addition to the interaction between two lateral spins, the exchange between two spin sites for an unpaired electron localized either on the lateral or the central sulfur of the molecule **I** is possible. Our calculations, however, show that in order to reach the values of the measured components of g -tensor the exchange should occur with the exchange frequency $\nu_{\text{ex}} = 3 \cdot 10^8 \text{ s}^{-1}$ and should lead to the narrowing of X , Y and Z spectral components up to 80, 60 and 0.3 mT, respectively. These values are much less than the linewidths obtained in experiment. Besides, the spin localization inside the lateral sulfuric group should lead to $\langle g \rangle \approx 2.0325$ in this case. This value seems to be too high for a sulfur containing radical. The average g -factor for the sample **I** is in the range of the values for RCCSS^+CCR and RCCSS^+CCR cation radicals, $2.014 \leq \langle g \rangle \leq 2.017$ [2, 10, 11] but exceeds that of tetrathiafulvalene derivatives, $2.011 \leq \langle g \rangle \leq 2.014$ [12, 13], in which an unpaired electron is localized near four central sulfur atoms of the TTF group. This additional circumstance confirms the supposition of the localization of non-interacting spins on the lateral trisulfur groups of the molecule **I**.

The excitation energies of the electron from the ground orbit to π^* and σ^* orbits ($\Delta E_{n\pi^*}$ and $\Delta E_{n\sigma^*}$, respectively) can be calculated from the measured g_{xx} and g_{yy} components of the g -tensor using the equation [14]

$$\Delta E_{nj} = \frac{g_e \rho_s \lambda_s}{g_{ii} - g_e} , \quad (5)$$

where $g_e = 2.00232$ is the g -factor for the free electron, ρ_s is the spin density on each lateral sulfur atom, and λ_s is the constant of spin-orbit coupling for sulfur. Using $\rho_s = 1.0$ and $\lambda_s = 41$ meV [14], $\Delta E_{n\pi^*} = 2.74$ and $\Delta E_{n\sigma^*} = 3.95$ eV were obtained for radical **I**. These values are close to the energy distances between the level occupied by the unpaired electron and nearest two excited levels (2.93 and 3.56 eV) calculated for $\overline{\text{HCS}\ddot{\text{S}}\text{SCH}^+}$ [2]. This is the evidence for the effective localization of an unpaired electron near one sulfur atom. The canonic components of the g -tensor of the radical **I** decrease down to $g_{xx} = 2.02977$, $g_{yy} = 2.02129$ and $g_{zz} = 2.00278$ when the sample is cooled down to 120 K probably because of the change of the molecular conformation and hence the increase of both ΔE_{nj} values as in some common solids. At the same time the linewidth of the Y and Z spectral components of the radical decreased down to 0.46 and 0.29 mT, respectively. This fact evidences that molecular σ motion freezes at cooling the system.

Figure 4 shows that the spectral canonic components of the trithiolium radical are broadened with the change of the molecular structure. This effect is accompanied by the shift of the components to the center of the spectrum weight because of the depinning of the spin (and hence charge) dynamics. The spectrum of the powder-like trithiolium salts may be considered as a superposition of spectra of separate radicals (spin-packets) with different orientation in the external magnetic field. The depinning of the molecule reorientation or/and spin diffusion leads to the change in the shape of the spectral lines similarly as it occurs at frequency exchange between different radical states. The rotation of the whole molecules is frozen and only a intramolecular spin motion or a libration of molecule segments with the rate $\nu_{||}$ is possible. It was shown earlier [12] that the shift of g -factor of organic π -conjugated systems arises mainly due to intramolecular spin diffusion. Therefore, $\nu_{||}$ can be determined from the Heisenberg equation by using the sum shift of the spectral components registered at the Δg_{ii} position relative to g_e and the modified equation [15]

$$\nu_{||} \cong \sum_i^3 \Delta g_{ii} \mu_B B_0 h^{-1} . \quad (6)$$

The rates of intramolecular spin diffusion in the samples under study are presented in Table 1.

The averaged EPR absorption linewidth at half-height depends mainly on spin-spin relaxation rate T_2^{-1} . For spin dipole interaction this value can be written in general as follows:

$$\gamma_e \langle \Delta B_{1/2} \rangle = T_2^{-1} = (T_2^{-1})_0 + (T_2^{-1})_1 + (T_2^{-1})_2 \quad , \quad (7)$$

where $(T_2^{-1})_0$ is attributed to the effect of the local magnetic fields induced by N neighboring spins. The $(T_2^{-1})_1$ term appears due to spin hopping between the molecules with the rate ν_1 , and the $(T_2^{-1})_2$ term is introduced in order to consider the spin diffusion near other paramagnetic centers with anisotropic magnetic parameters with the rate $\nu_{||}$.

The first term in Eq. (7) is determined by the second momentum M_2 of the Lorentzian line with Gaussian edges as $(T_2^{-1})_0 = \gamma_e (M_2/3)^{1/2}$. For equal paramagnetic centers chaotically situated and magnetically diluted in solid lattice this value can be calculated as [7]:

$$M_2 = \frac{3\pi}{10} \left(\frac{\mu_0}{4\pi} \right)^2 \gamma_e^2 \hbar^2 N \sum_k \frac{1 - 3 \cos^2 \theta}{r_{ik}^3} \quad , \quad (8)$$

where θ is the angle between the direction of \mathbf{B}_0 vector and radius-vector \mathbf{r}_{ik} , which connects the spins. Assuming for the simplicity that spin is situated in arbitrary sites of a cubic lattice with a constant $r_0 = ((N_c/3)^{-1/3})/2$ (here N_c is the concentration of the trithiole groups in the sample) so that the equality $\sum (1 - 3 \cos^2 \theta) r_{ik}^{-3} = 2.6 r_0^{-3}$ [16] holds, the second momenta of the samples were calculated (see Table 1).

The above mentioned line broadening means that additional channels for spin relaxation appear in samples II–IV compared with the sample I because of the spin motion. It may be either spin hopping between the molecules, intramolecular diffusion or librations of the spin together with a section of the molecule on which it is localized. This motion induces dipolar and/or hyperfine fields fluctuating rapidly with time at the other electron spin position. An auto-correlation function of this fluctuating field can be interpreted as a sum of various frequency components from dc to infinite. The spin flip leads to a resonance (Larmor) frequency absorption and therefore to the change in both lattice temperature and spin relaxation [17]. As the space coordinates of the spins precessing in different local magnetic fields are changed by the spin motion, the interaction of such centers is equivalent to the intrinsic chaotic dynamics in the system [7]. It takes a possibility to determine the other terms of Eq. (7) using the relations for relaxation rates of the spins moving in low-viscous systems as applied to discrete electron spin system. Therefore the spin hopping between the molecules should accelerate spin-spin relaxation by the value [18]

$$(T_2^{-1})_1 = \frac{1}{10} \left(\frac{\mu_0}{4\pi} \right)^2 \gamma_e^4 \hbar^2 S(S+1) \times \sum_k \frac{(1 - 3 \cos^2 \theta)^2}{r_{ik}^6} (6J(0) + 10J(\omega_e) + 4J(2\omega_e)) , \quad (9)$$

where $J(\omega_e) = (\nu_{\perp} + \omega_e^2/\nu_{\perp})^{-1}$ is the motional spectral density function at the operation frequency $\omega_e = 2\pi\nu_e$. Note that the analogous functional form was used by Mizoguchi in the study of spin dynamics in solid polycrystalline fullerenes [19].

The influence of the anisotropy of the g -factor on spin relaxation is described by the term [18]

$$(T_2^{-1})_2 = \frac{(g':g')\mu_B^2 B_0^2}{60\hbar^2} (8J(0) + 6J(\omega_e)) , \quad (10)$$

where $(g':g') = g_{xx}^2 + g_{yy}^2 + g_{zz}^2 - 3\langle g \rangle^2$ is the square of the g -tensor components averaged over all molecule orientation and $J(\omega_e) = (\nu_{\parallel} + \omega_e^2/\nu_{\parallel})^{-1}$.

The rates of the spin motion does not exceed operation frequency $\omega_e \gg \nu_{\parallel,\perp}$, so $J(\omega_e) = J(2\omega_e) = 0$ in Eq. (9) and $J(\omega_e) = 0$ in Eq. (10). Therefore the rate of spin hopping between the molecules can be calculated using $J(0)\sum(1 - 3\cos^2\theta)r_{ik}^{-6} = \pi N/45DR$ [18] (here $D = \nu_{\perp}r_0^2/6$ is the intermolecular spin diffusion coefficient and R is the radius of molecule) in Eq. (9) and $J(0) = \nu_{\parallel}^{-1}$ [20] in Eq. (10). The values of ν_{\perp} estimated for samples I–IV from the experimental data and from Eqs. (8)–(10) are summarized in Table 1.

In order to analyze the electron transport in the investigated systems, the limitations for electron transfer should be taken into account. The spin in the molecule I can diffuse along the π -conjugated system to the TTF unit. This should be accompanied by a decrease in its g_{xx} value down to 2.014–2.011 [12, 13]. This means the energy barrier higher than 4 eV (see Eq. (5)) exists for this spin transfer. Besides, SR groups form an additional barrier in such kind of π -conjugated system. Both lateral S atoms of the molecules possess a spin that prevents an intermolecular spin hopping. It leads to the freezing of the spin motion in this system. In the case of the molecule II, there are no appropriate vacancies for spin and hence, charge hopping from one internal site to another. Therefore, the intermolecular spin/charge dynamics are comparatively slow in this system. The unpaired electron and charge localized on the trithiolium group of the molecule II can take part mainly in the intramolecular libration motion of the group. This hypothesis is confirmed only by a shift of the spectral component X to the center of weight of the spectrum (Fig. 4). One and two above mentioned inside vacancies exist in the molecules III and IV, respectively. Besides, both spin and charge can hop from trithiolium group of these molecules to the same site of the neighboring molecules. This leads to an acceleration of both intra- and intermolecular spin/charge transfer and an in-

crease of anisotropy of such motion $\nu_{\parallel}/\nu_{\perp}$ from approximately 20 in the sample **II**, to 30 in the sample **III**, and to 300 in the sample **IV** (see Table 1). The anisotropy of the spin motion in the sample **III** is lower compared with that in the sample **IV** caused by the existence of $-\text{SCH}_3$ substituents in benzene ring, which build an energy barrier for this process. It seems that if ν_{\parallel} is the rate of libration of trithiolium group in the system **II**, it is the effective rate of libration and electron transfer in the systems **III** and **IV** with the predominance of electron diffusion in the latter. If one takes into account that the linewidth is defined mainly by an intramolecular spin motion with the effective activation energy E_a , then $E_a \approx 4$ meV can be evaluated for this motion in the samples **III** and **IV**. This value lies in the range of typical activation energies (1–10 meV) of electron transfer in various π -conjugated organic substances [21]. However it should be emphasized that such processes are not accompanied by any intramolecular reorganization, e.g., disruption of an S-S bond which requires $E_a > 1$ eV.

4. Conclusions

We showed that ac susceptometry and EPR spectroscopy are powerful tools for elucidation of electronic and magnetic properties of trithioles. The analysis of magnetic resonance data allowed us to investigate separately intramolecular spin dynamics and intermolecular spin transfer. The rates of these processes and their anisotropy depend on the structure of molecules and their packing.

Electron transport in organic substances is very interesting not only from experimental but also from theoretical point of view. In order to clarify intramolecular electron transfer the magnetic and relaxation properties of trithiolium radicals in solutions of different polarity may be investigated by X-band and also by high resolution D-band ($\nu_e = 140$ GHz) EPR spectroscopy. Further experiments are in progress and will be discussed in future publications.

References

- [1] Cameron T.S., Haddon R.C., Mattar S.M., Parsons S., Passmore J., Ramirez A.P.: *J. Chem. Soc. Chem. Commun.* **1991**, 358–360.
- [2] Cameron T.S., Haddon R.C., Mattar S.M., Parsons S., Passmore J., Ramirez A.P.: *J. Chem. Soc. Dalton Trans.* **1992**, 1563–1572.
- [3] Chenard B.L., Harlow R.L., Johnson A.L., Vladuchick S.A.: *J. Am. Chem. Soc.* **107**, 3871–3876 (1985)
- [4] Fanghanel E., Herrmann R., Naarmann H.: *Tetrahedron* **51**, 2533–2542 (1995)
- [5] Vonsovskii S.V.: *Magnetism*, p. 107. Moscow: Nauka 1971.
- [6] Blakemore J.S.: *Solid State Physics*, p. 526. London: Cambridge University Press 1985.
- [7] Lebedev Ya.S., Muromtsev V.I.: *EPR and Relaxation of Stabilized Radicals*, p. 33. Moscow: Khimija 1972.
- [8] Herrmann R.: Ph. Dr. Thesis, Martin-Luther-Universität Halle-Wittenberg, Merseburg, 1995.
- [9] Parmon V.N., Kokorin A.I., Gidomirov G.M.: *Stable Biradicals*. Moscow: Nauka 1980.
- [10] Russel G.A., Tanikaga R., Talaty E.R.: *J. Am. Chem. Soc.* **94**, 6125–6129 (1971)

- [11] Bock H., Rittmeyer P., Krebs A., Schultz K., Voss J., Kopke B.: *Phosphorus Sulfur* **19**, 131–136 (1984)
- [12] Krinichnyi V.I.: *2-mm Wave Band Spectroscopy of Condensed Systems*. Boca Raton: CRC Press 1995.
- [13] Williams J.M., Ferraro J.R., Thorn R.J., Carlson K.D., Geiser U., Wang H.H., Kini A.M., Whangbo M.-H.: *Organic Superconductors (Including Fullerenes): Synthesis, Structure, Properties and Theory*. Englewood Cliffs: Prentice-Hall 1992.
- [14] Buchachenko A.L., Wasserman A.M.: *Stable Radicals*, p. 92. Moscow: Khimija 1973.
- [15] Poole Ch.P.: *Electron Spin Resonance*, p. 466. London: Int. Sci. Publ. 1967.
- [16] Losche A.: *Kerninduktion*. Berlin: VEB Deutscher Verlag der Wissenschaften 1957.
- [17] Redfield A.: *Phys. Rev.* **98**, 1787–1794 (1955)
- [18] Carrington A., MacLachlan A.D.: *Introduction to Magnetic Resonance*, p. 230. New York: Harper and Row 1967.
- [19] Mizoguchi K.: *J. Phys. Chem. Solids* **354**, 1693–1698 (1993)
- [20] Abragam A.: *The Principles of Nuclear Magnetism*. London: Oxford University Press 1961.
- [21] Silinsh E.A., Kurik M.V., Chapek V.: *Electronic Processes in Organic Molecular Crystals: The Localization and Polarization Phenomena*, p. 329. Riga: Zinatne 1998.

Author's address: Prof. Dr. V. I. Krinichnyi, Institute for Chemical Physics, Russian Academy of Sciences, Chernogolovka, 142432 M.D., Russian Federation