A double-frequency signal appeared at $T < T_N$ at a magnetic field intensity H corresponding to excitation of the oscillations of the first branch. No frequency doubling was observed in the paramagnetic region.

Figures 1 and 2 show the temperature dependences of the double-frequency signal amplitudes for $MnCO_3$ and $CsMnF_3$. A sharp increase of the amplitude and a change of the signal phase by π were observed for both investigated antiferromagnets following smooth variation of the temperature in the vicinity of $T = T_{br}$, corresponding to biresonant frequency doubling.

The magnitude and form of the "biresonance" peak agree with the theory if the experimental values are used for the widths of the resonance lines and for the temperature dependence $\nu_2(T)$.

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INFLUENCE OF ROTATIONAL DIFFUSION ON THE PARAMAGNETIC RESONANCE LINE SHAPE

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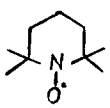
The EPR spectrum of paramagnetic particles in the dilute state in viscous media is determined by the degree of averaging, by the rotational motions of the particles (with a correlation time τ_c), of the anisotropy of the electronic Zeeman (tensor g_{ik}) and hyperfine (tensor f_{ik}) interactions [1]. Usually the anisotropy of the interactions is characterized by the parameter

$$\sigma = \max\left\{H_0\left|\beta\left(g_{zz} - \frac{g_{xx}+g_{yy}}{2}\right)\right|,\quad \left|f_{zz} - \frac{f_{xx}+f_{yy}}{2}\right|\right\},$$

where β is the Bohr magneton, H_0 the constant magnetic field, and g_{ij} and f_{ii} the principal values of the tensors.

Although it is of considerable interest (for example, for the study of high-molecular compounds [2]), there is no exact theory of EPR, and the experimental data are very scanty outside the region $\sigma\tau_c < 1$ where perturbation theory holds (fast rotations). To calculate the line shape in the region of slow rotations, $\sigma\tau_c \geq 1$, it is necessary to specify a concrete model of the random process [3, 4]. We shall show that the EPR line shape in a viscous medium can be well described by a theory that regards rotational motions as random changes of the molecule orientation by finite angles.

We chose as the paramagnetic particle the imino-acid radical [5, 6]



The unpaired electron in such radicals is localized mainly at the nitrogen nucleus. To simplify the calculations, we used a radical with the rare isotope N^{15} (nuclear spin I = 1/2).

The spin Hamiltonian of the electronic interaction in the laboratory frame, the z' axis of which coincides with the direction of the field H_0, can be written in this case in the form [1]

$$\mathcal{H} = \beta g_{z'k'} S_{k'} H_0 + \hbar f_{i'k'} S_{i'} I_{k'},$$

where S and I are the electron and nuclear spin operators (i', k' = x', y', z').

The rotational diffusion of particles was described by a stationary purely-discontinuous random Kolmogorov-Feller process [7] under the additional assumption that the change of orientation through any angle is equally probable. A method developed in [8] was used to obtain for the magnetic moment of the system $\vec{M}(\tau, \Omega_0)$, averaged over the random "trajectories" with fixed initial orientation Ω_0 (Ω is the set of Euler angles), the following expression:

$$\frac{\partial M(\tau, \Omega_0)}{\partial \tau} + \frac{1}{\tau_0}\left[M(\tau, \Omega_0) - \int M(\tau, \Omega_0)\frac{d\Omega_0}{8\pi^2}\right] + \frac{i}{\hbar}\left[M(\tau, \Omega_0), \mathcal{H}\right] = 0, \qquad (1)$$

where τ_0 is the time between the jump-like changes of the orientations, $\int \vec{M}(\tau, \Omega_0) d\Omega_0/8\pi^2$ is the value of the magnetic-moment operator in the Heisenberg representation, averaged completely over the random process, and $[\vec{M}(\tau, \Omega_0), \mathcal{H}]$ is the commutator.

The relaxation function is given by [9]

$$G_{x'x'} = Sp \int M_{x'}(\tau, \Omega_0) M_{x'} d\Omega_0$$

where $M_{x'} = (\beta/\hbar) S_{x'}$ is the magnetic-moment operator in the Schrodinger representation and Sp denotes summation over the spin variables. The absorption line shape is the Fourier transform of the relaxation function.

For the concrete calculations we used the parameter values $g_{xx} = 2.089$, $g_{yy} = 2.0061$, $g_{zz} = 2.0027$, $f_{zz} = 8.0 \times 10^8$ sec^{-1}, $f_{xx} = f_{yy} = 1.4 \times 10^8$ sec^{-1}, and $H_0 = 3.300$ Oe; this yields $\sigma \approx 7 \times 10^8$ sec^{-1} and an observation frequency $\omega_0 = 5.8 \times 10^{10}$ sec^{-1}.

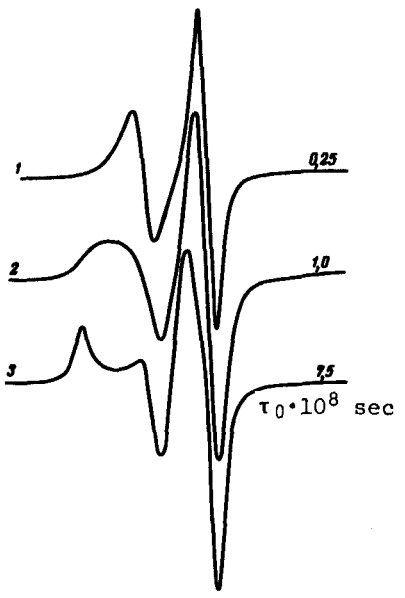


Fig. 1

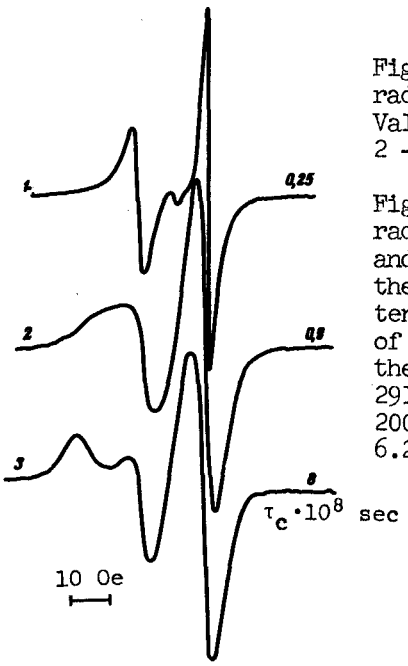


Fig. 2

Fig. 1. Theoretical EPR spectra of radical at different values of τ_0. Values of $\sigma\tau_0$ on plots: 1 - 1.7, 2 - 7, 3 - 52.

Fig. 2. Experimental spectra of radical in a solution of glycerine and 1.72% of H_2O. Spectrum 1 shows the line of the isotope N^{14} (content in sample $\sim$ 10%). The values of the temperature (T, °K) and of the viscosity (η, poise) are: 1 - 291, 7.0; 2 - 278, 23.5; 3 - 262.5, 200. The values of $\sigma\tau_c$ are 1.7, 6.2, and 56, respectively.

An exact solution of (1) was found for $\mathcal{H} = \mathcal{H}_{sec}$ ($\mathcal{H}_{sec}$ is that part of the Hamiltonian which commutes with S_z,). This approximation is valid when $(\omega_0\tau_0)^2 >> 1$ ($\tau_0 \geq 5\times10^{-11}$ sec). Theoretical EPR spectra were constructed for the entire interval of partcical interest 10^{-10} sec $\leq \tau_0 \leq 10^{-7}$sec ($0.07 \leq \sigma\tau_0 \leq 70$). Examples of the derivatives of the absorption lines in the region of slow rotations are shown in Fig. 1.

We investigated experimentally magnetically-dilute solutions of the radical in mixtures of glycerine and water, the viscosity of which was measured independently. Examples of the derivative of the EPR line shape (λ = 3 cm) of the radical at different temperatures are shown in Fig. 2.

To identify the experimetal spectra, we used the Stokes-Einstein formula customarily employed in the theory of magnetic relaxation [10], derived within the framework of hydrodynamic concepts

$$\tau_c = \frac{4\pi a^3}{3kT}\eta \ ,$$

where η is the viscosity of the medium and a is the effective radius of the radical. In the region of fast rotations ($\tau_c < 10^{-9}$ sec) τ_c was determined independently from the with of the resonance lines [11]. This has made it possible to calculate the hydrodynamic radius of the radical (a = 1.6 ± 0.1 Å. The values of τ_c corresponding to the experimental spectra in the region of slow rotations (Fig. 2) were calculated with the aid of formula (2), using the known radius and viscosity.

The theoretical and experimental spectra practically coincide, making it possible to conclude that the rotational diffusion can be effectively described by the relatively simple model of the purely-discontinuous random process. However, the fact that the parameters τ_0 and τ_c coincide within the limits of experimental error (~15%) for identical spectra raises the question of the possible equivalence of the models of jumplike and continuous diffusion for the description of paramagnetic-resonance phenomena.

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DEFLECTION OF POWERFUL LIGHT BEAMS UNDER THE INFLUENCE OF WIND IN ABSORBING MEDIA

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The subject of the present communication is a discussion of the effect of lateral wind on the propagation of a powerful laser beam under conditions of thermal self-action in absorbing media. As is well known, heating of the medium by the light beam gives rise to lens feffects, such as thermal self-focusing or self-defocusing. The removal of heat by the wind from the beam region changes the thermal regime in the medium, and hence also the properties