

The lower two transitions are of first order, while the upper has features of both first- and second-order transitions; however, judging from thermo-optical measurements, it is apparently also of first order, but close to second order.

Crystals with  $x < 0.5$  reveal in the upper ferroelectric phase, in a field  $E_y$ , double electric-hysteresis loops of a peculiar nature, corresponding to the induction of a ferroelectric phase I in the ferroelectric phase m (i.e., to the appearance of a Y component of the spontaneous polarization  $P_s$ ).

3. The character of the phase diagram and the independent behavior of the components of  $P_s$  (Fig. 2) allow us to draw a conclusion, compatible with the symmetry, that the Y component of  $P_s$  and the m component of  $P_s$  perpendicular to it (lying in the m-plane of the para-phase) not only have different microscopic mechanisms, but are also independent of each other. It is probable that the m component of  $P_s$  and accordingly the upper phase transition are due to ordering of hydrogen bonds, while the Y component of  $P_s$  and the two lower phase transition are due to a shift of the  $Na^+$  ions and a distortion of the  $SeO_3^{2-}$  groups. This hypothesis, which was advanced by us earlier [6], agrees well with the thermo-optical measurement data. (In crystals with  $x < 0.5$ , obviously, this hypothesis can also be verified directly, say by the NMR spectroscopy method.)

Thus, crystals of the system  $Na(D_xH_{1-x})_3(SeO_3)_2$  have an unusual isotopic effect, and apparently constitute the first example of ferroelectrics having spontaneous-polarization components that are independent and differ in their nature.

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#### FREE RADICALS AS CHAINS OF SPINS WITH ANTIFERROMAGNETIC INTERACTION

Yu. S. Karimov

Branch of Institute of Chemical Physics, USSR Academy of Sciences

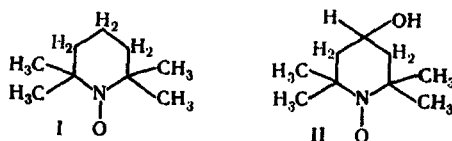
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There are few experimental studies of linear chains of spins. This is due primarily to the lack of suitable objects. There is only one known compound,  $Cu(NH_3)_4SO_4$ , for which a linear structure has been proposed [1]. From the theoretical point of view, the one-dimensional model is of considerable interest. This is connected with the fact that such a model admits of an exact solution under certain simplifications [2,3], and can also be calculated

with good approximation in a rather general case [4, 5].

Earlier investigations of a series of iminoxyl radicals by the EPR method have revealed that the magnetic susceptibility of two of them



passes through a maximum at liquid-helium temperatures [6]. This was not connected with the transition of the samples into an antiferromagnetic state, since the EPR signal was observed at all temperatures. In the present study, we investigated these two radicals at lower temperatures and in magnetic fields up to 60 kOe. The infralow temperatures were obtained by the adiabatic-demagnetization method. The magnetic fields needed for the demagnetization and for the measurement of the moment of the sample were produced with superconducting solenoids. We investigated single-crystal samples weighing 15 - 20 mg with good faceting.

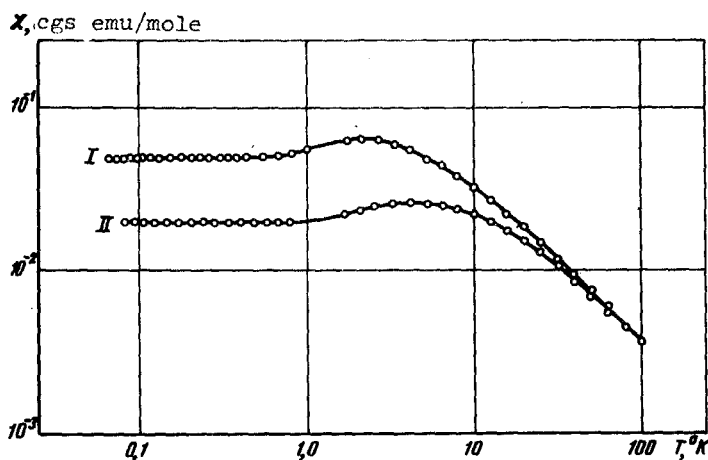


Fig. 1. Temperature dependence of magnetic susceptibility

Figure 1 shows the temperature dependence of the magnetic susceptibility for the two samples. With decreasing temperature, the susceptibility reaches a maximum, after which it decreases by approximately 25% and remains constant. No dependence of the susceptibility on the single-crystal orientation was observed. We plotted the field dependence of the magnetic moment for both radicals. At liquid-helium temperatures, the moment increases linearly with the field, the deviations from nonlinearity not exceeding 10% at 1.7°K. At lower temperatures, these deviations remain small for the radical II, but a strong increase of the magnetic moment is observed for radical I in strong fields (Fig. 2).

Such a behavior of the paramagnet is well described by the Heisenberg one-dimensional model with isotropic antiferromagnetic interaction [5]. All the physical characteristics of

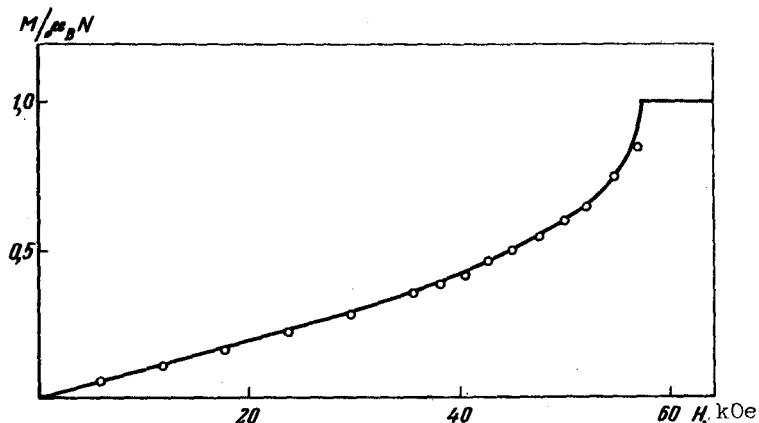


Fig. 2. Field dependence of the magnetic moment of sample I at  $T = 0.07^\circ\text{K}$

the paramagnet are functions of one parameter - the exchange integral  $J$ . The value of  $J$  can be determined most accurately from the  $M(H)$  dependence. The solid line in Fig. 2 shows the theoretical dependence, taken from [5] at  $T = 0$ . The abscissa scale is chosen such as to make the experimental values fit the curve best. A phase transition should be observed at  $T = 0$  and in a field  $H_{\text{cr}} = 2J/\mu_B$ . There is no transition if  $0 < T < J/k$ , the curve has no kink, but its form changes only in a narrow field region near  $H_{\text{cr}}$ . It can therefore be concluded from Fig. 2 that  $2J/\mu_B = 57 \pm 1$  kOe or  $J = 2.64 \times 10^{-16}$  erg for the radical I.

Another quantity characterizing a linear chain of spins is the magnetic susceptibility at  $T = 0$ . This quantity is connected with the exchange integral by the relation [5]

$$4J\chi_0/Ng^2\mu_B^2 = 0.259.$$

If we use the obtained value of  $J$  and the measured value of the susceptibility  $\chi_0 = 49.7 \times 10^{-3}$  cgs emu/mole, then we get for the radical I

$$4J\chi_0/Ng^2\mu_B^2 = 0.253.$$

The agreement between experiment and theory is quite good.

For the radical II, the value of  $J$  can be estimated from the temperature at which the susceptibility goes through a maximum [4, 5]:

$$J \approx 0.85kT_{\text{max}} = 6.0 \times 10^{-16} \text{ erg.}$$

It is clear that a similar increase of the magnetic moment near  $H_{\text{cr}}$  should be observed in much stronger fields. Thus, organic paramagnets are convenient objects for the study of the magnetic behavior of linear structures.

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Line 5 from the bottom, reads "deviations from nonlinearity,.." should read "deviations from linearity..."