

transitions in ruby at  $T = 77^\circ\text{K}$  (the spin-lattice relaxation time  $\tau_1 = 50 \mu\text{sec}$ , the lifetime of the level  $\bar{E}$  is  $\sim 5 \text{ msec}$ ), we can show that it is impossible to obtain a steady-state negative temperature between the levels  $E_1$  and  $E_2$ . The negative temperature observed in our experiments, and the generation associated with it, are caused by nonstationary processes. Calculations show that generation cannot proceed for longer than  $2-3 \tau_1$ .

In the ideal case, if the spin-lattice relaxation process is neglected, it is possible to find the maximum energy that our three level system is able to radiate: it is  $Nh\nu/6$ . For our specimen with a chromium ion concentration of  $\sim 0.05\%$ , this energy is  $\sim 2.5 \text{ erg}$ . Since generation lasts  $\sim 150 \mu\text{sec}$ , the maximum radiation power should not exceed  $1.7 \text{ mW}$ . The generated power observed in the experiment indicates the large role of relaxation processes.

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### COEXISTENCE OF ANTIFERROMAGNETIC AND SPECIAL DIELECTRIC PROPERTIES IN THE $\text{BiFeO}_3$ - $\text{LaFeO}_3$ SYSTEM

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THE compound  $\text{BiFeO}_3$  is ferroelectric with a Curie temperature  $T_C \sim 850^\circ\text{C}$  and antiferromagnetic with type G magnetic structure and Neel point  $T_N \sim 380^\circ\text{C}$ .<sup>[1-4]</sup> The attention of a number of investigators<sup>[5,6]</sup> has been turned recently toward a new class of substances combining ferroelectric and special magnetic properties and thereby having great scientific and practical interest. Complex investigations (x-ray, dielectric, and magnetic) of binary systems of compounds which to some degree or other combine special dielectric and magnetic properties are of great value for a thorough study of these phenomena.

The present paper describes results of such an investigation of the system  $\text{BiFeO}_3$ - $\text{LaFeO}_3$ , in

which  $\text{BiFeO}_3$  is a ferroelectric antiferromagnet, and  $\text{LaFeO}_3$  is antiferromagnetic with weak ferromagnetism.<sup>[7]</sup> The usual ceramic techniques were used to synthesize the samples. X-ray analysis of the samples at room temperature showed that a complete series of solid solutions with the perovskite structure is formed over the entire concentration range. These solid solutions exist in four modifications, depending on the  $\text{LaFeO}_3$  content (in mole percent): up to 18.8—rhombohedral; from 18.8 to 55—pseudomonoclinic I (PMI); from 55 to 73—pseudomonoclinic II (PMII); above 73—pseudomonoclinic III (PMIII).

The magnetic measurements were carried out by the Faraday method from room temperature to  $500^\circ\text{C}$  in fields up to 8 kOe. The samples of all compositions demonstrated unusual magnetic properties.

Figure 1 shows the temperature dependence of the magnetization  $\sigma(t)$  for different samples of the system in a field of 7600 Oe. Similar results for the magnetic susceptibility of  $\text{BiFeO}_3$  as a function of temperature have been obtained by Smolenskiĭ et al.<sup>[8]</sup>

In Fig. 2, besides the lines of the phase transition in this system (of which more will be said below), is presented the dependence of the room-temperature spontaneous magnetization  $\sigma_0$  on the composition of the samples.

The presence of spontaneous magnetization in the compositions with 12.5, 15, and 17.5%  $\text{LaFeO}_3$ , as well as in all investigated samples in the three pseudomonoclinic regions and the similarity between the magnetization vs. temperature curves for these compositions and the like curves for the weak ferromagnet  $\text{LaFeO}_3$ , is evidence that all these samples are also antiferromagnets with weak ferromagnetism.

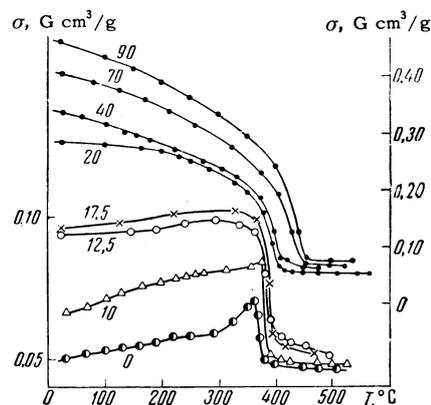


FIG. 1. Temperature dependence of the specific magnetization of samples of the system  $\text{BiFeO}_3$ - $\text{LaFeO}_3$  in a field of 7600 Oe. The numbers on the curves indicate the  $\text{LaFeO}_3$  content in mole %. The scale on the right is for compositions containing 20, 40, 70, and 90 mole %  $\text{LaFeO}_3$ .

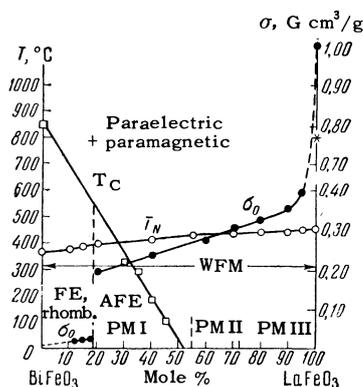


FIG. 2. Phase diagram of the system  $\text{BiFeO}_3\text{-LaFeO}_3$  and the dependence of the spontaneous magnetization at  $20^\circ\text{C}$  on the composition of the samples (FE – ferroelectric region, AFE – antiferro-electric, WFM – weakly ferromagnetic); \* – approximate value of the spontaneous magnetization of  $\text{LaFeO}_3$  at  $20^\circ\text{C}$  from the data of [9].

The spontaneous magnetizations of the sample of 10%  $\text{LaFeO}_3$  and of pure  $\text{BiFeO}_3$  were found to be commensurate within the limits of experimental error— $(8-9) \times 10^{-3} \text{ G cm}^3/\text{g}$ . However, because of the presence of a sharp maximum in the temperature dependence of the magnetization for these compositions and the existence of spontaneous magnetization in samples of the rhombohedral modification, to which belong also both the 10%  $\text{LaFeO}_3$  and pure  $\text{BiFeO}_3$ , we conclude that these samples also are weakly ferromagnetic.

At the boundary between the rhombohedral and PMI modifications there is a step-like increase in the spontaneous magnetization. With further increase in the  $\text{LaFeO}_3$  content up to 90%  $\sigma_0$ , to a first approximation, increases linearly and continuously. A detailed study of the behavior of the spontaneous magnetization at the boundaries between PMI and PMII and between PMII and PMIII was not made, since our main interest was in the region of concentrations up to  $\sim 50\%$   $\text{LaFeO}_3$ .

A sharper increase in the magnitude of the spontaneous magnetization is found to occur in the region of concentrations from 90 to 100%  $\text{LaFeO}_3$ .

In addition to the x-ray and magnetic measurements, investigations of the temperature dependence of the dielectric constant were carried out on a number of samples whose conductivity was not too high for the measurements to be made.

On the basis of the totality of the results of all these measurements a phase diagram of the system was constructed (Fig. 2). The line  $T_C$  (Curie temperature) is the line for the phase transition from a ferro- or antiferroelectric state to a paraelectric one, and the line  $T_N$  (Néel point) is the line for the transition from a state with antiferromagnetic ordering to a paramagnetic state.

The following very interesting regions can be distinguished in the phase diagram: up to 18.8%  $\text{LaFeO}_3$  the ferroelectric properties are combined with weak ferromagnetism, and in the region from 18.8 to 55%  $\text{LaFeO}_3$  antiferroelectricity coexists with weak ferromagnetism, and the coexistence of electrical and magnetic ordering takes place in a wide interval of temperatures higher than room temperature. It should be emphasized that the sharp rise in spontaneous magnetization at the boundary between the rhombohedral and pseudo-monoclinic I modifications with the transition from the ferroelectric to the antiferroelectric phase seems to us to be not accidental, but indicative of some interaction between the special dielectric and special magnetic states.

It is natural to expect coupling, since both characteristics depend strongly on the very same factor—the interatomic spacings. The existence of this coupling was theoretically predicted by Smolenskii, [10] and the experimental results cited earlier [5,6] are evidence for it.

A more detailed exposition of the results of the x-ray, dielectric, and magnetic investigations and a discussion of them will be given in the journal "Kristallografiya" (Soviet Phys. Crystallography).

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<sup>10</sup>G. A. Smolenskii, *FTT* 4, 1095 (1962), *Soviet Phys. Solid State* 4, 807 (1962).