

G. A. Solenkii, E. I. Golovenchits, and V. A. Sanina. *Magnetic phase transition induced by high-power optical pumping.* A magnetic phase transition in a magnetically ordered crystal, stimulated by high-power optical pumping, has been observed for the first time. The effect was produced in the antiferromagnet  $\text{EuCrO}_3$  ( $T = 180$  K). The  $\text{Eu}^{3+}({}^7F_0)$  ions that were nonmagnetic prior to the introduction of the pump are found to occupy a state with a magnetic moment ( ${}^7F_1$ ) when the pump is turned on, and take part in the exchange interaction with the  $\text{Cr}^{3+}$  ions and, through them, with each other. The result of this is that long-range antiferromagnetic order with ordering temperature  $\sim 100$  K appears in the subsystem of the  $\text{Eu}^{3+}$  ions.

The phase transition was detected experimentally by investigating the homogeneous ( $k = 0$ ) antiferromagnetic resonance (AFMR) in the Cr-subsystem. The resonance AFMR field of the Cr-subsystem changes discontinuously when the pump is turned off. Thereafter, over a period of several minutes, it continues to shift toward the stable value which remains unaltered so long as the low temperature is maintained. The effect has a threshold character both in relation to the pulse power and the duration of the pump. Two situations can be realized for crystals of different thickness, depending on the duration and power delivered by the pump: the new AFMR signal coexists with the old (initial) signal, or one observes a single new AFMR signal (if the crystal thickness does not exceed  $100 \mu\text{m}$  at the highest pump power). As the temperature is increased, the effect is found to vanish for  $T \geq 100$  K.

The effect can be explained in terms of the model of coupled exchange polarons. In their excited state, the  $\text{Eu}^{3+}({}^7F_1)$  ions have a magnetic moment and, since they are located in the magnetically ordered lattice of  $\text{Cr}^{3+}$

ions, they take part in the exchange interaction with them, which in turn gives rise to the appearance of the exchange polarons. When several neighboring  $\text{Eu}^{3+}$  ions are excited at the same time, they interact with one another through the  $\text{Cr}^{3+}$  lattice (indirect f-d-f exchange) which leads to a reduction in the energy of this cluster. It then turns out that for clusters consisting of  $N = 4, 8, 12$ , etc., coupled polarons, the energy is lower than for clusters containing one polaron less or more. Such clusters turn out to be metastable, since their decay along the single-ion channel is energetically unfavorable, whereas decay in to all the ions simultaneously has low probability. When the lifetime of such metastable clusters is greater than the separation between the pump pulses, their number will increase while the pumping radiation is present, and a macro-region containing magnetized  $\text{Eu}^{3+}$  ions will appear. The model predicts that when the number of excited  $\text{Eu}^{3+}$  ions in a cluster reaches a certain value ( $N \geq 24$ ), its energy becomes negative, i. e., the exchange reduction in energy compensates the splitting between the ground and the excited states of  $\text{Eu}^{3+}$  ( $E = {}^7F_1 - {}^7F_0 \cong 300 \text{ cm}^{-1}$ ). The crystal state with the magnetized  $\text{Eu}^{3+}$  ion becomes the ground state. Spontaneous transition of the  $\text{EuCrO}_3$  crystal to the state with the magnetic Eu-subsystem does not occur at low temperatures because the probability of the simultaneous thermal excitation of several neighboring  $\text{Eu}^{3+}$  ions to the  ${}^7F_1$  level is low. At high temperatures, on the other hand, when this process is possible, the exchange interaction is absent.

It follows from the model that the f-d-f exchange reaches its maximum in the course of antiferromagnetic ordering of the Cr host. The Eu-subsystem is then also in the antiferromagnetic ordering of the Cr host.

The Eu-subsystem is then also in the antiferromagnetic state with the antiferromagnetic axis lying along the  $Y$  axis of the crystal. Measurements have been carried out of the magnetic moment of the crystal as a function of temperature for the principal crystal directions before and after exposure to the high-power optical pump. Magnetic measurements confirm the presence of antiferromagnetism in the Eu-subsystem after exposure to the pumping radiation along the  $Y$  axis.

Measurements have also been carried out of the optical absorption spectra of  $\text{EuCrO}$  in the range 2200–500 nm at temperatures between 4.2 and 300 K before and after exposure to the optical pump. The absorption spectra recorded after the application of the pump contain an absorption band with a maximum at 1400 nm, and a number of other features connected with appearance of magnetic order in the Eu-subsystem, but these features vanish on heating ( $T \geq 140$  K). We note that the absorption spectra recorded before the pump was turned off contained features that had not been seen previously in magnetic crystals based on the  $\text{Cr}^{3+}$  ions, namely, a giant red shift of the electronic vibrational band  ${}^4T_2$  ( $\sim 3000 \text{ cm}^{-1}$ ) and an anomalous behavior of the  $R$ -lines of  $\text{Cr}^{3+}$ . These features are due to the fact that, in the  ${}^4T_2$  band and in  $\text{EuCrO}$ , the excitation of the

$\text{Cr-Eu}$  exciton is energetically favorable. This exciton takes the form of an excited  $\text{Cr}^{3+}$  ion (in the  ${}^4T_2$  band), surrounded by excited  $\text{Eu}^{3+}({}^7F_1)$  ions. Polarization of the magnetic moments of the  $\text{Eu}^{3+}$  ions as a result of f-d-f exchange produces a substantial reduction in the energy of this exciton. A number of independent optical effects were used to estimate the model parameters: the magnitude of f-d-f exchange and the position of the first excited state of  $\text{Eu}^{3+}$ . These were found to agree with one another and with the data obtained as a result of magnetic and resonance measurements.

It is thus clear from resonance, magnetic, and optical data that high-power optical pumping gives rise to a magnetic phase transition in  $\text{EuCrO}_3$ . The experimental data confirm the basic assumptions of the theoretical model put forward to describe the phenomenon. The basic results are published in the following papers.

<sup>1</sup>E. I. Golovenchits, V. A. Sanina, and T. A. Shaplygina, *Zh. Eksp. Teor. Fiz.* **80**, 1911 (1981) [*Sov. Phys. JETP* **53**, 992 (1981)].

<sup>2</sup>E. I. Golovenchits and V. A. Sanina, *Fiz. Tverd. Tela (Leningrad)* **24**, 375 (1982) [*Sov. Phys. Solid State* **24**, 213 (1982)].