Long-lived, magnetically ordered state in $EuCrO_3$ excited by optical pumping

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A long-lived, magnetically ordered state in $EuCrO_3$ produced as a result of exchange interaction of optically excited Eu^{3+} ions, which have a magnetic moment and a magnetically ordered chromium matrix, was observed experimentally. This effect is explained in terms of the model for bound magnetic polaritons.

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A strong influence of the $3d$-$4f$ exchange on the frequency of the antiferromagnetic resonance (AFMR) of a chromium subsystem has been observed in the investigation of magnetic resonance in $ErCrO_3$.[1] In $EuCrO_3$ the Eu^{3+} ground-state ions are nonmagnetic.[2,3] In the excited states, however, they have a magnetic moment. If the lifetimes of such excited states are sufficiently large, then a shift of the AFMR line in the Cr subsystem is expected to occur as a result of optical excitation of the Eu ions.

The AFMR was investigated in $EuCrO_3$ by simultaneous generation of the millimeter waves (1.0–1.5 mm) and a powerful pulsed optical pumping (532 nm). The heating of the samples was controlled by a carbon resistor and a thermocouple, and did not exceed 1 K at 5 K.

At a certain threshold pulsed pumping power of sufficient duration, we observed a new resonance absorption signal. This signal was shifted relative to the original AFMR (prior to pumping) in the direction of large fields. The signal remained for many hours. The dependence of the signals on pumping and their time behavior are shown in Figs. 1 and 2. We shall mention the more characteristic experimental observations. 1) The original AFMR signal of the Cr subsystem either disappeared after maximum pumping with respect to power and time (Fig. 2) or greatly decreased its intensity and shifted slightly toward the large fields (Fig. 1). 2) A relatively fast increase of the intensity of the new signal and a decrease of its resonance field were observed during the first minutes after the laser was turned off. Subsequently, the intensity of the signal did not vary with time, and the resonance field decreased more slowly (Fig. 2). Figure 3 shows the temperature dependences of the resonance fields of the original AFMR signal and of the new signal. It can be seen that the signals

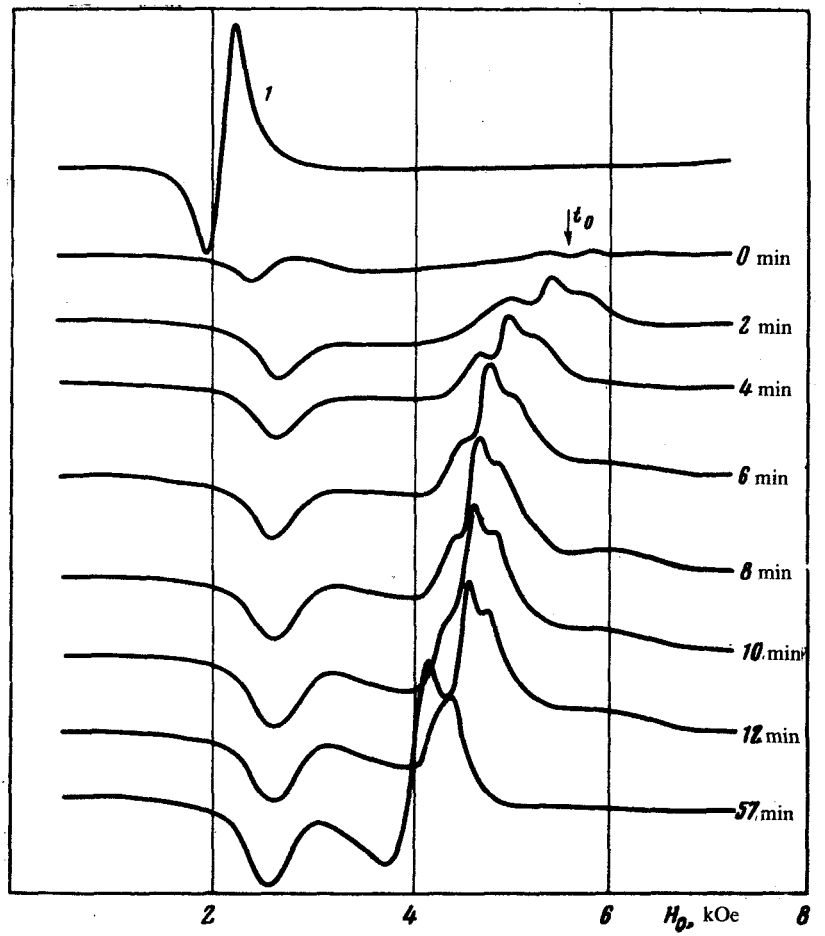

FIG. 1. Traces of the AFMR lines in $EuCrO_3$. Scanning of the magnetic field is plotted along the X axis and the intensity of the signals is plotted along the Y axis. The sample is $2\times2\times0.15$ mm^3, $T = 5$ K, and $f = 210$ GHz. Line 1 represents AFMR of the chromium subsystem prior to pumping. The following curves were recorded during pumping and after the laser was turned off. The parameters of the optical pumping are: pulse power $P_p = 180$ kW, pulse duration $\tau_p = 15$ nsec, pulse repetition frequency $\Omega = 12.5$ Hz, and duration of pumping $t = 1.5$ min.

combine beginning at $T \sim 90$ K. 4) We investigated the effect of the more powerful and longest pumping on the AFMR in $YCrO_3$. We found no evidence of the effect. There were also no anomalies due to optical pumping of pure $EuAlO_3$ and with a small concentration of Cr impurity.[4] We can infer from this that the effect is due to the interaction of Eu and Cr ions.

Since the resonance field is shifted and the intensity of the new signal does not decrease as a result of the time relaxation, we conclude that the new signal is an AFMR signal of the Cr subsystem, which is shifted relative to the original signal. The shift is determined by the effective field produced as a result of interaction of the ions

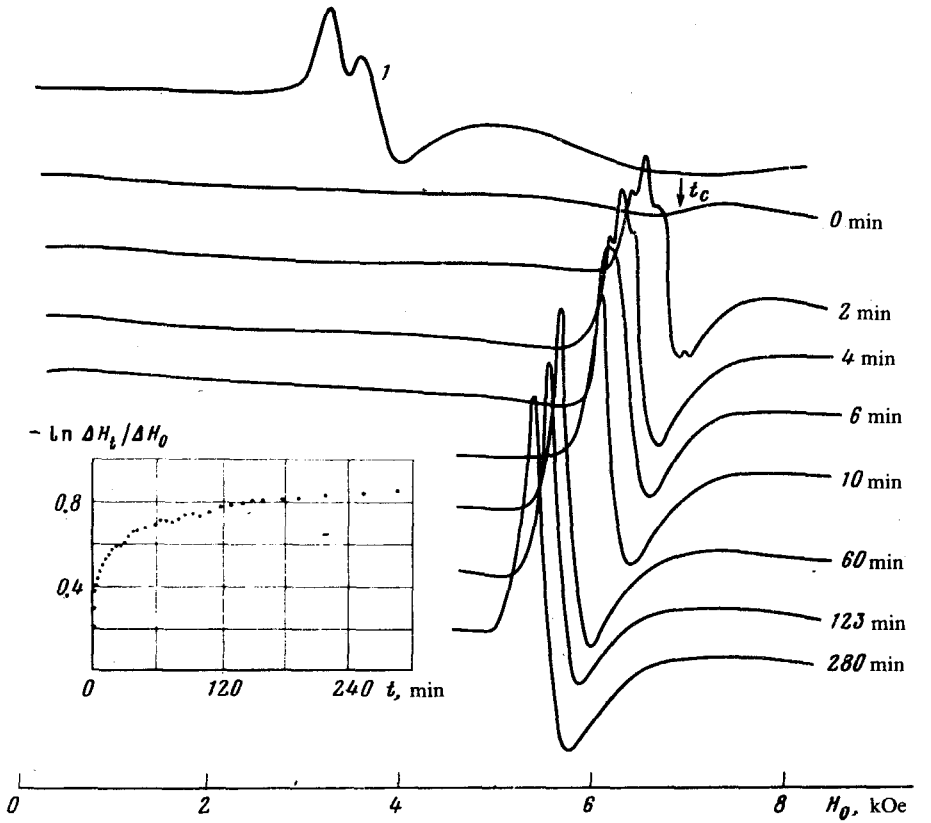

FIG. 2. The same as in Fig. 1 but for the pumping parameters: $P_P = 250$ kW, $\tau_p = 15$ nsec, $\Omega = 25$ Hz, and $t = 4$ min. The lower left-hand side shows the time dependence of $\ln(\Delta H_t/\Delta H_0)$, where $\Delta H_{0,t}$ is the difference between the resonance field of the observed signals and H_{res} of the initial AFMR at the times $t = 0$ and t, respectively.

of the Cr subsystem with the excited Eu^{3+} ions that have a magnetic moment. The temperature dependence also indicates this. In fact, at $T \sim 90$ K the new signal combines with the original AFMR signal, and in the low-temperature region the dependence is analogous to that observed in the crystals with magnetic rare-earth ions in the ground state.[1]

Thus, to explain the experimental data, we must assume that the excited Eu ions have a magnetic moment whose lifetime is extremely long. The explanation of the anomalously large lifetime should be sought in the exchange interaction of the excited Eu ions with magnetically ordered matrix, which was not observed in the ground state of these ions. In the state with a magnetic moment, Eu^{3+} polarizes the matrix, which reduces its energy, i.e., a "magnetic polaron" is produced. The existence of a power threshold and the need for a sufficiently long pumping leads us to believe that the long-lived state is a collective state. The long lifetime of the excited states can be accounted for qualitatively if we assume that the energy of the state comprised of N

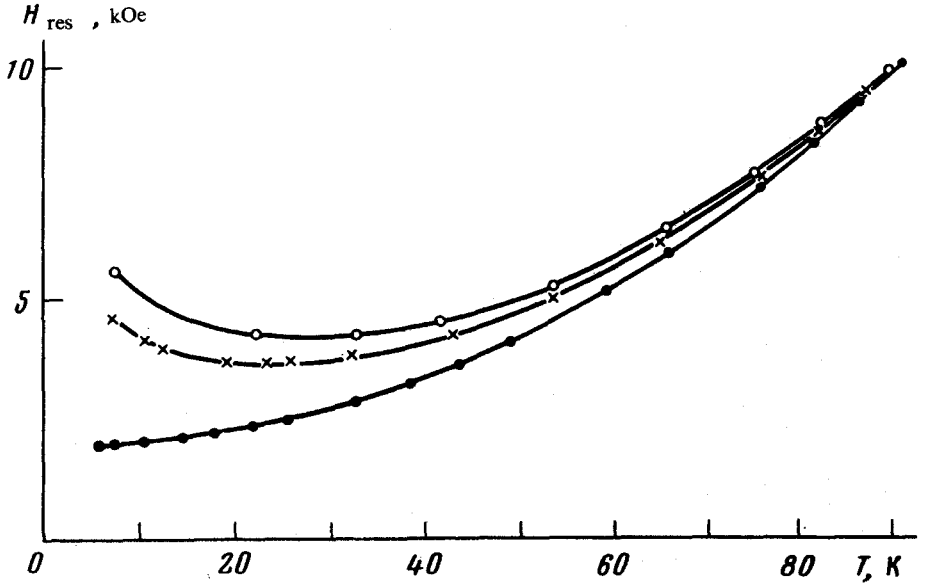

FIG. 3. Temperature dependences of the resonance fields for: ●, initial AFMR without pumping, O,new signal at the moment the laser was turned off, and ×, the same signal one hour after the laser was turned off.

bound polarons is lower than that comprised of $N-1$ bound polarons. In fact, the transition to the ground state of one Eu^{3+} ion in this case is energetically disadvantageous, and that of many ions simultaneously is unlikely. Thus, the occurrence of the effect only at sufficiently large pumping power is attributable to the necessity for a simultaneous excitation of several closely spaced Eu ions. With the appearance of a magnetic-polaron cluster, which can survive the dead time between the pulses, the polarons begin to accumulate in proportion to irradiation, which shifts the resonance line.

We shall demonstrate that the energy can decrease with increasing number of polarons, on the basis of the model described by the Hamiltonian

$$\mathcal{H} = N\epsilon + \sum_{ij} a_2(\mathbf{r}_i - \mathbf{r}_j) I_i S_j + \mathcal{H}_s. \tag{1}$$

Here N is the number of excited Eu^{3+} ions and ϵ is the energy of the excited ion, disregarding the exchange. The second term describes the $3d$-$4f$ exchange, where I and S are the operators of the Eu and Cr magnetic moments, respectively. $\mathcal{H}_s$ is the Hamiltonian of the chromium matrix.

We shall assume that the Eu ions are in a self-consistent field of the chromium matrix whose state in turn is determined by the states of the Eu ions. Thus, the effective Hamiltonian of the Eu ions can be written as follows:

$$\mathcal{H}_{eff} = N\epsilon + \sum_{ij} a(\mathbf{r}_i - \mathbf{r}_j) \langle S_j \rangle I_i - \tfrac{1}{4} \sum_{ij} b_{\alpha\beta}(\mathbf{r}_i - \mathbf{r}_j) I_i^{\alpha} I_j^{\beta}. \tag{2}$$

Here $\langle S_j \rangle$ is the average spin of the Cr ions.

$$b_{\alpha\beta}(\mathbf{r}_i - \mathbf{r}_j) = \frac{v_0}{g^2\mu^2} \sum_{kl} a(\mathbf{r}_i - \mathbf{r}_k)\, \chi_{\alpha\beta}(\mathbf{r}_k - \mathbf{r}_l)\, a(\mathbf{r}_l - \mathbf{r}_j), \tag{3}$$

where v_0 is the volume per chromium atom and $\chi_{\alpha\beta}(\mathbf{r}_k - \mathbf{r}_l)$ is the susceptibility of the chromium matrix.

In our case, the second term in Eq. (2), which is nonvanishing solely due to a weak ferromagnetism, is disregarded. The main term in Eq. (2) is the third term which describes the effective exchange between the Eu^{3+} ions via the chromium matrix. Disregarding the longitudinal susceptibility of Cr and the transverse anisotropy, we can reduce Eq. (2) to the Hamiltonian of the X-Y model. We take into account in this Hamiltonian only the self-action and the interaction of the nearest neighbors. The corresponding exchange constants are $b_0 = 0.46\, a^2/|A|$ and $b_1 = 0.87\, a^2|A|$. A is the exchange constant of the Cr ions.

If the excited states of Eu^{3+} are the lowest $(^7F_1)$[1)] states, we obtain the following ground-state energies of one, two, three, and four bound polarons: $E_1 = \epsilon - b_0$; $E_2 = 2(\epsilon - b_0) - 2.2b_0$; $E_3 = 3(\epsilon - b_0) - 4.0b_0$; $E_4 = 4(\epsilon - b_0) - 8.0b_0$. These values fit into the following scheme: each polaron has the energy $(\epsilon - b_0)$ and each bond has $(-2b_0)$. An extrapolation of this systematic feature to $N \gg 1$ bound polarons gives $E_N = N(\epsilon - 7b_0)$.

It can be seen from these results that $E_1 > E_2$ when $\epsilon < 3.2b_0$, $E_2 > E_3$ when $\epsilon < 2.8b_0$, and $E_3 > E_4$ when $\epsilon < 5b_0$. It ϵ corresponds to the $^7F_1$ state (300 cm^{-1}) and b_0 is of the order of 90 K (Fig. 3) when the effect vanishes, then the condition $E_3 > E_4$ is certainly fulfilled.

We note that in this case $\epsilon < 7b_0$ and at sufficiently large N, $E_N < 0$, i.e., the state comprised of N bound polarons is the ground state of the crystal. The probability for a spontaneous transition to this state is very small since it requires a simultaneous excitation of N Eu^{3+} ions. The lifetime of the state without the polarons relative to such transition is of the order of $\tau(\epsilon\hbar^{-1}\tau)^N$, where τ^{-1} is the width of the $^7F_1$ level.

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[1)]The specially set up experiments, which will be discussed in depth in another paper, favor the assumption that the $^7F_1$ states are the long-lived states.

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