Oscillation of the sign of the Faraday rotation caused by Eu^{3+} ions in an ultrastrong magnetic field up to 11 MG

A. I. Pavlovskiĭ, V. V. Druzhinin, O. M. Tatsenko, N. P. Kolokol'chikov, A. I. Bykov, and M. I. Dolotenko

Moscow Engineering-Physics Institute

(Submitted April 11, 1980)

Pis'ma Zh. Eksp. Teor. Fiz. **31**, No. 11, 659–653 (5 June 1980)

The Faraday effect (FE) was investigated in a plastic containing Eu^{3+} ions. The FE caused by the Eu^{3+} ions changes its sign twice in 5 and 11-MG fields. This is due to competition of the paramagnetic and intrinsic diamagnetic moments.

PACS numbers: 78.20.Ls, 75.20. – g

The MK-1-type explosion magnetic generator was described in Ref. 1. This device is capable of producing pulsed magnetic fields up to ≈ 10 MG. The method of measuring the FE using this apparatus was described in a previous paper.[2] In this letter we describe an investigation of the FE in a plastic containing Eu^{3+} ions with a concentration 4×10^{20} cm^{-3}. The field was measured from an oscillogram of the FE in TF-5 glass ($l = 2.07$ mm, Verdet constant $V = 0.0457$ min/cm G) at a 632.8-nm wavelength. The FE of a 0.67-cm-long test sample was measured at this frequency at room temperature. The oscillograms for both samples are shown in Fig. 1. The Verdet constant of a plastic without impurities is $V_{\rm plas} = 0.0097$ min/cm Oe, in which the FE can be assumed to be linear. The nonlinearity of the FE in transparent glasses is determined by the equation $V = a - b/B^2$, where $b\cdot B^2/a \approx 10^{-17} B^2$, which gives a contribution of less than 1% in 10^7-G fields.[3] Thus, by subtracting the matrix rotation angle $\theta_{\rm plas}$ from the total rotation angle of the sample, we determine $\theta(Eu^{3+})$—the contribution of the Eu^{3+} sublattice. The dependence of $\theta(Eu^{3+})$ of B in Fig. 2 shows that the initial negative angle increases to ≈ 2.5 MG, after which it decreases, changes its sign, passes through a maximum in ≈ 9-MG field, and again changes its sign in ≈ 11-MG field.

This field dependence is attributable to the fact that the Eu^{3+} $(4f^6)$ ion in the ground state, which has an antiparallel arrangement of the orbital and spin magnetic moments (the $^7F_0$ state), is a van Vleck paramagnet because of the first excited state $^7F_1$ with an energy of 310 cm^{-1}.[8] The total shift of the levels in ≈ 10-MG field is $\approx 10^3$ cm^{-1}, i.e., it would seem that the Zeeman sublevels of the $^7F_1$ multiplet in ≈ 4 to 5-MG field cross the ground-state level and the ion acquires a normal paramagnetism. This, however, does not occur because of the interaction of the levels with a different J but the same M. A diagonalization of the 49th-order matrix

$$\langle SLJM| H_{S_\theta} + \mu B(\hat{L}_2 + 2\hat{S}_2)|SLJ'M\rangle = E(J)\,\delta_{J;J'} + \mu B$$

$$\times (-1)^{J-M}\begin{pmatrix} J & 1 & J' \\ -M & 0 & M \end{pmatrix} \sqrt{(2J+1)(2J'+1)}\,(-1)^{L+1}\left[(-1)^{J+S}\begin{Bmatrix} L & J & S \\ J' & L & 1 \end{Bmatrix}\right.$$

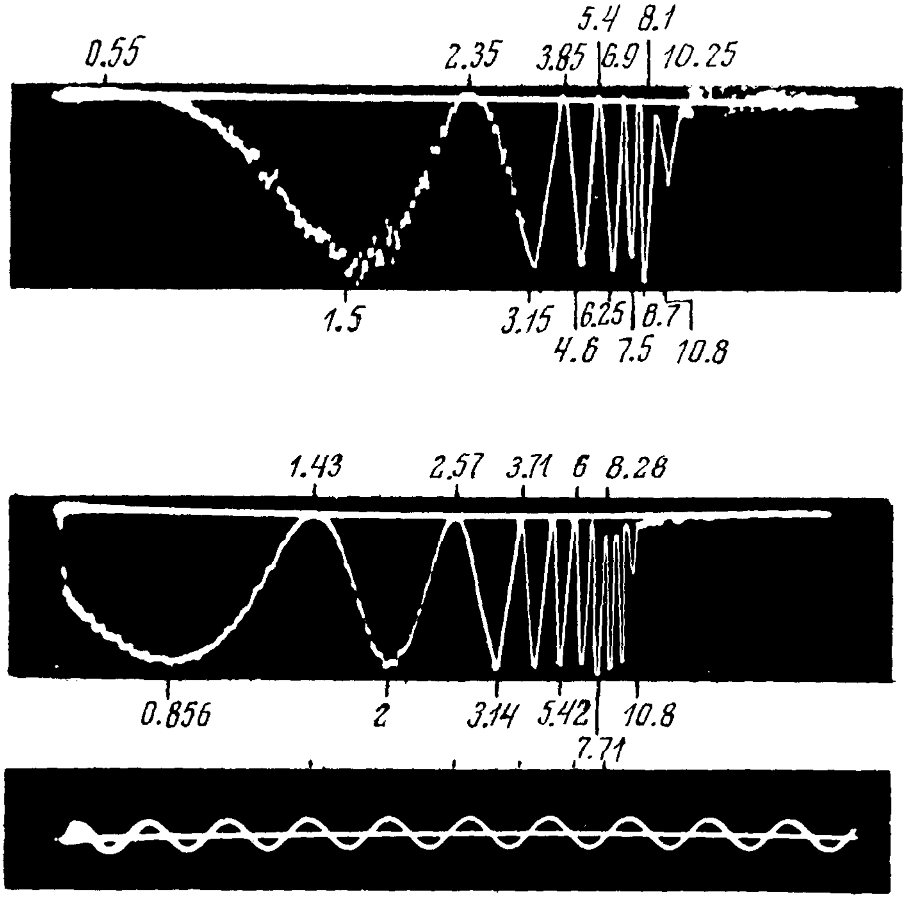

FIG. 1. Oscillograms of Faraday rotation: top, plastic with Eu^{3+} impurity; middle, flint glass; bottom, calibration sinusoid with 1-μsec period. The numbers denote the field in MG. The first mark for the plastic trace corresponds to a 45° angle, and for the TF-5 flint glass to a 130° angle. The oscillograms begin at 10 μsec. The distance between the nearest minima is 180°.

$$\times \sqrt{L(L+1)(2L+1)} + 2(-1)^{S+J'} \begin{Bmatrix} S & J & L \\ J' & S & 1 \end{Bmatrix} \sqrt{S(S+1)(2S+1)} \Bigg] \qquad (1)$$

shows that the $J = M = 0 - (00)$ level is strongly repulsed from the (10) level, so that the crossing (00) and $(1-1)$ is possible only in the fields $B \approx 13$ MG.

It is known that the intrinsic diamagnetism is typical of all systems, increases linearly with the field and in a very strong field it can become larger than the uncompensated orbital and spin magnetic moment that saturates. Such competition in the Eu^{3+} ion is facilitated by the small paramagnetic contribution, which is the cause of oscillation of the sign of $\theta(Eu^{3+})$ with increasing field. We represent $\theta(Eu^{3+})$ in the

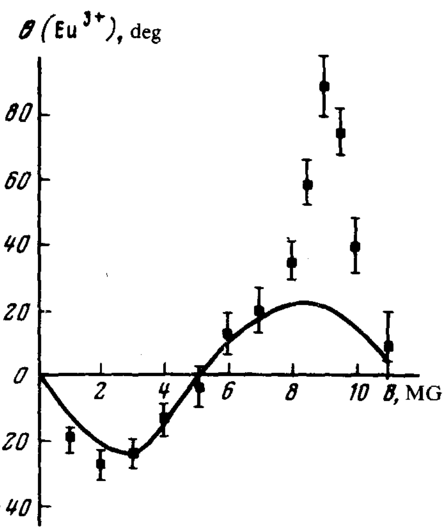

FIG. 2. Dependence of $\theta(Eu^{3+})$ on the magnetic-field induction. The points represent the experiment and the solid line denotes the calculation according to Eq. (2).

form of two terms: a negative paramagnetic term θ_p and a positive diamagnetic term θ_d, which are associated with the $^7F_1$ and $^7F_0$ polarization, respectively. In weak fields $|\theta_p| < \theta_d$ and $\theta(Eu^{3+})$ is negative. This is confirmed by the experimental data.[4] The field dependence of θ_p is determined by the proportionality $\theta_p \sim \bar{M}$, where $\bar{M}$ is the average moment of the excited state, and $\theta_d \sim \omega_0 B$, where ω_0 is the probability of finding the Eu^{3+} ion in the ground state. At $T = 300$ K $\omega_0 = 0.622$ in a weak field. As the field increases, $\bar{M}$ increases from -0.085 in a 1-MG field to -0.45 in a 11-MG field, i.e., it is not saturated, since the (1–1) magnetic level remains excited. In this case $d\bar{M}/dB \approx -0.085$ for $B \lesssim 3.5$, $d\bar{M}/dB \approx 0$ for $3.5 < B \lesssim 6$, and $d\bar{M}/dB \approx -0.032$ for $B > 6$ in units of MG^{-1} and MG, respectively, i.e., there are two linear regions separated by a plateau. As the field increases, ω_0 also increases because of the decrease in the effective degeneracy of the excited state, despite the approach of the $E(1-1)$ level toward $E(00)$. The largest value of $\omega_0 = 0.72$ is reached in a field $B \approx 5$ MG, after which ω_0 decreases linearly and approaches 0.5.

Thus, the sign reversal of $\theta(Eu^{3+})$ in $\approx$5-MG field is due to the largest value of ω_0 a temporary saturation of $\bar{M}$. The value of ω_0 decreases in $\approx$9-MG field and $\theta(Eu^{3+})$ again approaches a negative value. Quantitatively, this process is described by the equation

$$\theta(Eu^{3+}) = a\bar{M}l + \omega_0 V_d lB \,. \qquad (2)$$

The relationship between the two unknown parameters a and V_d can be obtained from the Verdet constant of $Eu(PO_3)_3$, after subtracting the contribution of $(PO_3)_3^{3-}$, which is known,[4,5] For weak fields this gives $\theta(Eu^{3+})/lB = -1.9\times 10^{-3}$ min/cm Oe. Using the ratio $a\bar{M}/B\omega_0 V_d = -1.78$, which typically corresponds to $V_p/V_d = -5$ for normal media, we can calculate $\theta(Eu^{3+})$ from Eq. (2). The results of the calculation and experimental data shown in Fig. 2 are in close agreement, except in the 8 to 10-MG region, where as appreciable quantitative discrepancy is observed. This discrepancy may be due to mixing of the $J\,0$ and $J\,-1$ wave functions of the analyzed states as well as the contribution of the high-frequency permeability.[6] We note that the temperature variation due to the magnetocaloric effect should not be noticeable ($\Delta T \lesssim 10$ K), since there is no level splitting or crossing[7] in the lower energy interval within the limits $kT = 207\ \text{cm}^{-1}$.

Thus, we have noticed that the intrinsic diamagnetism predominates over the uncompensated paramagnetism in an ultrastrong magnetic field.

[1]A. I. Pavlovskiĭ, N. P. Kolokol'chikov, M. I. Dolotenko, and A. I. Bykov, Prib. Tekh. Eksp. No. 5, 195 (1979).

[2]A. I. Pavlovskiĭ, N. P. Kolokol'chikov, V. V. Druzhinin, O. M. Tatsenko, A. I. Bykov, and M. I. Dolotenko, Pis'ma Zh. Eksp. Teor. Fiz. **30**, 211 (1979) [JETP Lett. **30**, 193 (1979)].

[3]V. V. Druzhinin and O. M. Tatsenko, Opt. Spektrosk. **36**, 733 (1974) [Opt. Spectrosc. (USSR) **36**, 426 (1974)].

[4]S. B. Berger, C. B. Rubinstein, C. R. Kurjian, and A. W. Treptow, Phys. Rev. A **133**, 723 (1964).

[5]O. M. Tatsenko, A. I. Pavlovskiĭ, and V. V. Druzhinin, Opt. Spektrosk. **42**, 147 (1977) [Opt. Spectrosc. (USSR) **42**, 81 (1977)].

[6]G. S. Krinchik and M. V. Chetkin, Zh. Eksp. Teor. Fiz. **36**, 1924 (1959) [Sov. Phys. JETP **9**, 1368 (1959)]; Usp. Fiz. Nauk **98**, 3 (1969) [Sov. Phys. Usp. **12**, 307 (1969–70)].

[7]V. V. Druzhinin, V. M. Mel'nikov, and V. V. Shkarubskiĭ, Fiz. Tverd. Tela **21**, 1750 (1979) [Sov. Phys. Solid State **21**, 1002 (1979)].

[8]J. D. Kingsley and J. S. Preher, Phys. Rev. **126**, 458 (1962).