CONFERENCES AND SYMPOSIA

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A scientific session of the Division of General Physics and Astronomy of the Russian Academy of Sciences was held on 25th February 1998 at the P L Kapitza Institute for Physical Problems, Russian Academy of Sciences. The following reports were presented at the session:

- (1) **Zvezdin A K, Lubashevskiĭ** I A (Institute of General Physics, Russian Academy of Sciences, Moscow), **Levitin R Z** (Moscow State University), **Platonov V V, Tatsenko O M** (Institute of Experimental Physics, Sarov). "Phase transitions in megagauss magnetic fields."
- (2) **Pustovoĭt V I** (Central Design Bureau of Unique Instrument Manufacture, Russian Academy of Sciences). "Collinear diffraction: phenomena and devices."

A brief presentation of the first report is given below.

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# Phase transitions in megagauss magnetic fields

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#### 1. Introduction

Using ultra-high magnetic fields for the investigation of new materials is a topical problem of present-day physics. The point is that such fields ( $\gtrsim 100~\text{T}$ ) as well as ultra-high pressures ( $\gtrsim 50~\text{GPa}$ ) and low temperatures ( $\lesssim 100~\text{K}$ ) cause an essential modification of the energy spectrum. This brings into existence new effects or nonlinear features in familiar phenomena, which provides a deeper insight into one or another property of solids.

This report reviews the results of recent research performed using MC-1 magneto-cumulative generators, which make it possible to obtain magnetic fields of up to  $10^3$  T in a volume of several cm<sup>3</sup> with high stability [1].

In fact, these magneto-cumulative generators realize the idea of producing ultra-high magnetic fields through the explosive compression of a magnetic flux. This idea was suggested by Sakharov in 1952. The basic elements of the MC-1 generator are a solenoid and a cylindrical charge. Schematically, the solenoid is a multifilar, multilayer coil wound by insulated copper wire. The outside and inside diameters of the solenoid are 152 mm and 140 mm respectively; the length is 300 mm. Such a solenoid lets a magnetic flux pass freely inwards and becomes conductive as the shock

wave induced by the explosion of a cylindrical charge passes through it. Ultra-high magnetic fields result in the following way. At first the discharge of a capacitor bank with an accumulated energy of 2 MJ brings into existence an initial magnetic field on the order of 20 T. Thereupon the solenoid is compressed by a cylindrical shock wave. As the shock wave passes through the solenoid, it welds the separate wires together into a compact conducting cylinder. Such a liner effectively captures and compresses the magnetic flux. Magnetic fields achievable under such conditions are measured using the Faraday effect.

This review presents the results of investigations into the properties of phase transitions in megagauss fields both in classic materials (ferromagnets, antiferromagnets) and in new ones, which have become the subject of much attention in recent years (nanostructured magnets, high-spin organometallic clusters, supramolecular systems). To be specific, let us consider separately three physical systems whose properties demonstrate the main characteristic features of phase transitions in such fields. Conceptually, these transitions are

- (1) first-order (discontinuous) phase transitions in rareearth metamagnets;
  - (2) second-order phase transitions in antiferromagnets;
  - (3) quantum transitions in mesoscopic magnetic clusters.
- It should be noted that the last problem is gaining crucial importance in the context of the development of a new line of investigation in materials science the creation of magnetic storage and recording media on the basis of individual nanometer-scale clusters and complex molecules.

## 2. Metamagnetic transitions in itinerant-electron ferrimagnets $RCo_2$

Phase transformations of magnetic structures are realized in multisublattice ferrimagnets and antiferromagnets. Of special fundamental and applied interest are high-anisotropic and hard ferrimagnetic materials, among them intermetallic ferrimagnetic materials of the R-T type, where R and T denote a rare-earth element and a transition metal of the Fe group, respectively. It is precisely these compounds that provide a basis for the search for new prospective materials for permanent magnets and information-recording systems with ultra-high writing density. The study of phase transitions induced by a magnetic field and their associated phase diagrams is a direct way to determine microscopic parameters and fundamental relationships.

The characteristic values of exchange fields in this class of substances are on the order of 100-400 T. Consequently, the critical fields of phase transitions lie in the range from 100 to 1000 T and can be measured using the ultra-high-field technique. At the present time, these materials are being investigated very actively, but only in the range of 10 to 50 T. Thus, the extension of these investigations into the region of

megagauss magnetic fields is a topical problem in the presentday physics of magnetic processes and may result in the discovery of fundamentally new effects and phenomena.

In the intermetallic compounds of rare earths and Fegroup transition elements, there coexist two electron subsystems. One of them is formed by the f electrons of a rareearth element, which are localized on atoms; the other consists of the hybridized d electrons of a transition and a rare-earth metal. The properties of the latter subsystem are usually described in terms of the band model of magnetism.

In the case of heavy rare earths, the f-d exchange interaction is negative and the magnetic moments of the rare-earth (f) and band (d) subsystems are aligned antiparallel to each other. Mathematically, these compounds may be treated as two-sublattice ferrimagnets. This picture is realized in RCo2 intermetallics (cubic Laves phase C15). Special interest in these materials is dictated by the fact that their band d subsystem is unstable, with its ground state being paramagnetic [2, 3]. In other words, in these materials the interaction of f electrons gives rise to spontaneous magnetization  $M_f$ , whereas the intrinsic interaction of d electrons is not too strong and, in the absence of an external magnetic field and d interaction, the f-d subsystem would be in the paramagnetic state. Nevertheless, an external field applied to the d subsystem will induce a preferential orientation of the magnetic moments of d electrons, and when its magnitude is on the order of  $H_M \sim 100$  T, the interaction of d electrons with each other will give rise to a new ferrimagnetic-type phase state of this subsystem. The possibility of such transitions being realized in itinerant magnets was first predicted conceptually by Wohlfarth and Rhodes [4]. This possibility is associated with some peculiarities of the energy dependence of the d electron density of states near the Fermi level (see, for example, [5]). In most RCo<sub>2</sub> compounds with magnetic rare earths, the d subsystem is in the ferromagnetic state, since the molecular field  $\lambda M_{\rm f}$  acting on it from the f subsystem ( $\lambda$  < 0 is the exchange coupling constant for the f-d exchange interaction) is greater than the metamagnetic transition field  $H_M$ .

Mathematically, the effective field  $\mathbf{H}_{\text{eff}}$  acting on the d subsystem in  $R\text{Co}_2$  compounds can be represented as a sum of the external  $\mathbf{H}$  and molecular fields:

$$\mathbf{H}_{\text{eff}} = \mathbf{H} + \lambda \mathbf{M}_{\text{f}} \,. \tag{1}$$

Therefore, when magnetizing these compounds, the net magnetic moment of the d subsystem is initially aligned antiparallel to the external magnetic field, since the effective field  $\mathbf{H}_{\mathrm{eff}}$  is negative, with the orientation of the external field taken as a positive direction. The magnitude of  $\mathbf{H}$  decreases with increasing  $\mathbf{H}_{\mathrm{eff}}$ , and when it reaches the value  $H_M$ , the d subsystem changes its state from ferrimagnetic to paramagnetic. The magnitude of the external field corresponding to this transition is

$$H_{\rm c1} = |\lambda| M_{\rm f} - H_M \,. \tag{2}$$

This implies that in such ferrimagnets a reentrant order—disorder transition from a magnetically ordered to a magnetically disordered paramagnetic state is possible.

With further increasing magnetic field, when its magnitude exceeds the value

$$H_{c2} = |\lambda| M_{\rm f} + H_M \,, \tag{3}$$

the d subsystem returns to the ferromagnetic state. However, the net magnetic moments of the d and f subsystems are now parallel and aligned with the external magnetic field. Thus, in ferrimagnetic RCo $_2$  compounds, there have to be observed two successive metamagnetic transitions associated with magnetization jumps during the processes of demagnetization and reentrant magnetization of the d subsystem in an external field.

In pure  $R\text{Co}_2$  compounds, these metamagnetic transitions have not been actually observed, since according to the estimates they would have to take place in magnetic fields in excess of 100 T. Nevertheless, metamagnetic transitions of this type have been realized experimentally and studied on substituted compounds  $(R,Y)(\text{Co},\text{Al})_2$  and  $(R,\text{Lu})(\text{Co},\text{Al})_2$ , where they are shifted into the region of lower fields [6, 7]. However, the phase diagrams of substituted compounds are more complex, since in these compounds metamagnetic transitions may 'interfere' with transitions into the non-collinear ferrimagnetic phase [7–9].

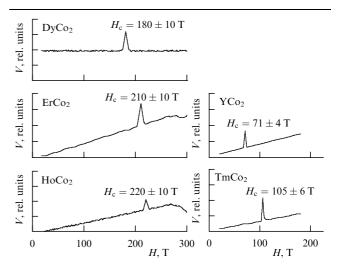
#### 2.1. Experiment

Measurements of the critical fields of metamagnetic transitions in the band subsystem of most  $R\text{Co}_2$  compounds involving heavy rare earths were performed at T=4.2~K using the induction method in pulsed magnetic fields up to 300 T [10]. The fields were obtained using the explosive technique [1]. The  $R\text{Co}_2$  compounds were used in the form of powders. The signal induced in the detecting coils can be written in the form

$$V(H) \sim K \frac{\mathrm{d}H}{\mathrm{d}t} + \frac{\mathrm{d}M}{\mathrm{d}t} = \left(K + \frac{\mathrm{d}M}{\mathrm{d}H}\right) \frac{\mathrm{d}H}{\mathrm{d}t}.$$

Since dH/dt is a monotonic function of the field, we can determine the magnitude of the metamagnetic transition field from the maximum of the signal V(H).

Figure 1 shows the signal induced in the detecting coils versus the magnetic field for some  $R\text{Co}_2$  intermetallics [10]. In all the curves, one can clearly see maxima that give an indication of a sharp increase in magnetization. The fields corresponding to these anomalies may be considered as the critical fields of the metamagnetic transitions. For nonmag-



**Figure 1.** Field dependence of the signal induced in the detecting coil of a pulsed solenoid for *R*Co<sub>2</sub> compouns at 4.2 K (from [10]).

netic YCo2 with only one band subsystem, the value of this field agrees with the  $H_M$  value obtained in [2] to within the experimental error. The critical fields  $H_{c1}$  or  $H_{c2}$  obtained for intermetallics with a magnetic rare earth are listed in Table 1. In addition, the magnitudes of these critical fields are also given that were calculated from the  $\lambda$  values obtained in indirect ways: from the Curie temperature of RCo2 compounds [11] and from the magnetization curves of substituted compounds  $(R,Y)(Co,Al)_2$  [6]. The correlation of these magnitudes with the experimental values of the critical fields has allowed the identification of the revealed transitions as the metamagnetic magnetization or demagnetization of the d subsystem. It is seen that there is only a qualitative agreement between the values of critical field calculated using data from [6, 11] and the experimental values. This indicates that the crystal-field effects in RCo2 which did not take the above into account are quite considerable, and additional investigations are required to improve this agreement.

Table 1. Critical fields of metamagnetic transitions in RCo<sub>2</sub>.

Rare- earth metal	$H_{c_1}$ , T			$H_{c_2}$ , T		
	Experi- ment	Theory	Theory [6]	Experi- ment	Theory [11]	Theory [6]
Tm		4	3; -12 [12]	105	144	143; 128 [12]
Er	55	83	123	210	223	263
Но	220	160	190		300	330
Dy	180	220	300		360	440

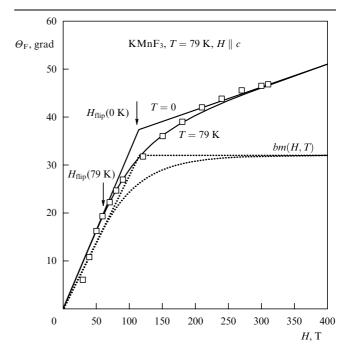
### 3. Spin-flip transition and Faraday rotation in the antiferromagnetic KMnF<sub>3</sub>

This section is concerned with manifestations of second-order phase transitions in megagauss fields. KMnF<sub>3</sub> is an adequate model object for revealing the relation between magnetooptical activity and the orbital moment of the excited states of quantum systems (magnetic ions). The  $Mn^{2+}$  ion belongs to an important class of ions which includes ions with a zero orbital moment in the ground state (Fe<sup>3+</sup>, Mn<sup>2+</sup>, Gd<sup>3+</sup>) and ions with a frozen orbital moment (Cr3+, Ni2+ in an octahedral crystal field). Magnetooptical activity of such quantum systems is determined by the orbital moments of the excited states and spin – orbit coupling. Faraday rotation measurements using ultra-high magnetic fields provide a good possibility for the study of this problem, owing to the competition of the ultra-high field with the 'spin-orbit' field  $H_{SL}$  in the vicinity of 120 T. The magnetooptics of quantum systems with a zero orbital moment are of great interest from a practical standpoint as well, since these ions enter into the composition of most magnetooptical materials.

#### 3.1. Experiment

The Faraday effect in KMnF<sub>3</sub> was measured at 79 K in pulsed magnetic fields up to 400 T produced by a magneto-cumulative generator (see, for example, [13]). A cylindrical sample of KMnF<sub>3</sub> 2.4 mm in diameter and 3.6 mm in thickness, cut along the c axis, was placed in a nitrogen cryostat. The measurements were conducted with laser radiation at a wavelength of 0.63  $\mu$ m. The light passed through a polarizer and analyzer and was then detected using two photomultipliers. One photomultiplier was used to detect Faraday rotation, the other to detect absorption.

The absorption of the sample was constant everywhere over the region under investigation. Giant values of the magnetic field were attained by explosive compression of the magnetic flux in the MC-1 generator. The magnetic field was measured using an inductive sensor (a coil) placed near the sample in liquid nitrogen. The experimental error did not exceed 5 %. An important feature of the magnetic-field dependence of Faraday rotation in KMnF<sub>3</sub> is the absence of saturation in fields up to 400 T, whereas the critical field of the spin-flip transition is on the order of 120 T at 0 K and 55 T at 79 K (Fig. 2).



**Figure 2.** Field dependence of Faraday rotation for a 3.8-mm thick  $KMnF_3$  sample ( $\Box$ , experiment; —, theory; dotted lines, the field dependence of the second term in (7) (that is, m).

#### 3.2. Magnetic structure and phase transitions

At room temperatures, KMnF<sub>3</sub> has a perovskite-type structure. As the temperature decreases to 184 K, a structural phase transition into the tetragonal phase  $(D_{2h}^{18})$  occurs [14], and then, at 81.5–88 K, into the orthorhombic phase  $(D_{2h}^{16})$  [15]. However, the distortions of the magnetic structure in passing from the simple axial ferromagnet to the slightly canted antiferromagnet are only on the order of 0.003° for the quantization angle [16].

The free energy of KMnF<sub>3</sub> can be represented in the form

$$\Phi = \Lambda M_1 M_2 - (M_1 + M_2) H + F_A - TS, \tag{4}$$

where  $M_1$  and  $M_2$  are the sublattice magnetizations,  $\Lambda$  is the exchange constant, S is the entropy; and  $F_A$  takes into account the magnetic anisotropy and antisymmetric exchange energy. In an ultra-high magnetic field the F term can be neglected, so that the magnetic structure of the crystal is determined by the competition of the exchange and Zeeman interactions. For the relative magnetization  $m = (M_1 + M_2)/2M_0$ , we have

$$m = \frac{H}{2H_E}$$
 at  $H < H_{\text{flip}}(T)$  (5)

and

$$m = B_J \left( \frac{\mu_0(H - H_E m)}{T} \right)$$
 at  $H > H_{\text{flip}}(T)$ . (6)

Here  $H_{\text{flip}} = 2H_E m_0(T)$  is the spin-flip transition field,  $m_0(T)$  is determined from the equation  $m_0(T) = B_J(T_N m_0/T)$ ,  $B_J(...)$  is the Brillouin function,  $H_E = \Lambda M_0 \approx 57$  T is the exchange field,  $\mu_0 = 5\mu_{\text{B}}$  is the magnetic moment of  $\text{Mn}^{2+}$ ,  $M_0 = \mu_0 N/2$ , and N is the concentration of  $\text{Mn}^{2+}$  ions. The corresponding magnetization curves at 0 K and 79 K are illustrated in Fig. 2 (dotted lines).

### 3.3. Magnetic field dependence of Faraday rotation $\Theta_{\mathrm{F}}$

Faraday rotation in KMnF<sub>3</sub> can be represented in the form [18]

$$\Theta_{\mathcal{F}}(H,T) = aH + bm(H,T), \qquad (7)$$

where a and b are constants, and m(H,T) is the relative magnetization determined by expressions (5) and (6). Using equation (7), the function  $\Theta_{\rm F}(H)$  was calculated. From a comparison of experimental data with calculated curves, the values of the constants were determined:  $a=0.048 \ {\rm deg} \ {\rm T}^{-1}$ ,  $b=32 \ {\rm deg}$  (Fig. 2).

Summing up the results of this part of our report, we can state the following. Faraday rotation in compounds with S ions, among them KMnF<sub>3</sub>, does not saturate in external magnetic fields up to 300-400 T. This fact is explained by the joint action of the effective spin-orbital field  $H_{SL}$  ( $\sim 100$  T), the exchange field ( $\sim 57$  T) and the external magnetic field  $H \lesssim 400$  T. A relatively small value of Faraday rotation in KMnF<sub>3</sub> at a wavelength of 0.63 µm may be attributed to nearly complete compensation for the negative diamagnetic contribution arising from the 6S-6P transitions by a positive paramagnetic contribution arising from the  $6S-4T_1(G)$  transition. In this situation, the KMnF<sub>3</sub> matrix essentially contributes to the Faraday effect.

## 4. High-spin nanoclusters $Mn_{12}Ac$ in megagauss fields

Mesoscopic-scale magnetic materials exhibit new and very interesting properties such as giant magnetostriction, magnetoresistance, the magnetocaloric effect, the existence of bistability on a cluster scale, and macroscopic quantum tunneling of magnetization [19–23]. In this respect molecular magnetic clusters involving transition-metal ions are of great interest. Such clusters can be considered an ideal model of a nanometer-size single-domain particle.

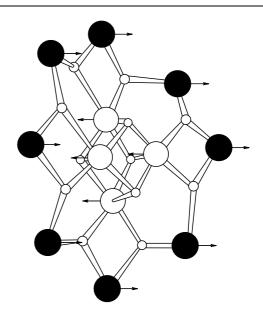
In most clusters exchange coupling between ions is antiferromagnetic, that is, the clusters can be considered as ferrimagnetic particles. The process of magnetization of such clusters may be effectively studied in only an ultra-high field, since the typical values of the exchange magnetic field in clusters are on the order of  $10^6$  G.

In this paper, one of the typical and most interesting highspin clusters,  $Mn_{12}Ac$ , is investigated. Its general formula is

$$[Mn_{12}(CH_3COO)_{16}(H_2O)_4O_{12}] \cdot 2CH_3COOH \cdot 4H_2O$$
.

The  $Mn_{12}Ac$  crystals are formed from individual  $Mn_{12}$  molecules, crystallization water, and acid molecules. The  $Mn_{12}$  molecules have a tetragonal symmetry; the unit cell volume is 3716 A . In each unit cell, twelve Mn ions form a cluster with four inner  $Mn^{4+}$  ions (spin S=3/2) surrounded

by eight  $Mn^{3+}$  ions (spin S=2) (Fig. 3). The  $Mn_{12}Ac$  cluster exhibits a Jahn-Teller-type transformation with a large distortion of the structure.



**Figure 3.** Schematic sketch of an  $Mn_{12}Ac$  cluster. The large circles correspond to magnetic Mn ions (four central  $Mn^{4+}$  ions and eight  $Mn^{3+}$  ions), the small circles correspond to oxygen atoms. Four water molecules and 16 acetic ligands are not shown.

Antiferromagnetic exchange coupling in the cluster makes its ground state stable with S = 10 in which the parallel spins of the Mn<sup>4+</sup> ions are aligned antiparallel to the spins of the Mn<sup>3+</sup> ions. An external magnetic field induces successive transitions from the ferrimagnetic phase into a noncollinear canted phase and then from the latter phase into the ferromagnetic phase. In the canted phase, the angle between sublattice magnetizations varies with the field. The spin reorientation of Mn ions in this cluster was shown to be similar to the magnetization process in a Néel-type ferrimagnet, on the one hand, but at the same time it exhibits a number of features inherent exclusively in quantum systems, on the other. At low temperatures, the spin reorientation is a sequence of quantum jumps, which become smoother with increasing temperature, and the magnetization curve eventually approaches its classical analogue.

#### 4.1. Theory

Exchange interactions between Mn ions are described by the Hamiltonian [24, 26]

$$H = \mu H \sum_{i=1}^{12} S_i - 2J_1(S_1S_2 + S_3S_4 + S_5S_6 + S_7S_8)$$

$$-2J_2(S_2S_9 + S_4S_9 + S_4S_{10} + S_6S_{10} + S_6S_{11} + S_8S_{11} + S_8S_{12} + S_2S_{12})$$

$$-2J_3(S_2S_4 + S_2S_6 + S_2S_8 + S_4S_6 + S_4S_8 + S_6S_8)$$

$$-2J_4(S_1S_9 + S_1S_{12} + S_3S_9 + S_3S_{10} + S_5S_{10} + S_5S_{11} + S_7S_{11} + S_7S_{12}),$$
(8)

where the first term describes the Zeeman interaction of the ions with an external magnetic field, and  $\mu = g\mu_B$  ( $g \approx 2$ ).

According to [24], the values of the exchange integrals are of the following orders:  $J_1 \approx -150 \text{ cm}^{-1}$ ,  $J_2 \approx J_3 \approx -60 \text{ cm}^{-1}$ ,  $|J_4| \leq 30 \text{ cm}^{-1}$ . The uniaxial-anisotropy energy (the z axis is assumed to be directed along the axis of symmetry of the molecule) is predominantly determined by the crystal field. An estimate of the dipolar interaction between clusters gives values of about 0.01 T. The energy levels of interest for study of the magnetization process were calculated in [26]. Going to dimensionless quantities  $\epsilon_i = E_i(h)/|J_1|$  and  $h = 2\mu_B H/|J_1|$ , one can represent the energy levels in the form

$$\epsilon_i(h) = \epsilon_i(0) + \delta \epsilon_i - h(i+9),$$
 (9)

where

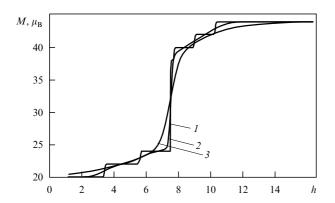
$$\epsilon_i(0) = 3i \sum_{k=1}^{5} \delta_{ki} + 5(i-2) \sum_{k=6}^{9} \delta_{ki} + 7(i-4) \sum_{k=10}^{13} \delta_{ki}, (10)$$

 $\delta_{ki}$  is the Kronecker symbol, and the values of  $\delta\epsilon_i$  are given in [26]. Formulas (9) and (10) determine the values of the critical fields  $h_{c1}$ ,  $h_{c2}$ ,  $h_{c3}$ , etc., which correspond to ground-level crossovers.

The field and temperature (at  $T \le 300$  K) dependence of magnetization for a Mn<sub>12</sub> cluster has the form [25]

$$M(h,\tau) = 2\mu_{\rm B} \frac{\sum_{i=1}^{13} i \exp[-\epsilon_i(h)/\tau]}{\sum_{i=1}^{13} \exp[-\epsilon_i(h)/\tau]} + 18\mu_{\rm B} \,,$$

where  $\tau = T/|J_1|$ , and  $\epsilon_i(h)$  are determined in (9). Figure 4 shows typical magnetization curves calculated according to this formula at  $J_1 = -150$  cm<sup>-1</sup>,  $J_2 = -60$  cm<sup>-1</sup>,  $J_3 = 60$  cm<sup>-1</sup>, and  $J_4 = 0$ .

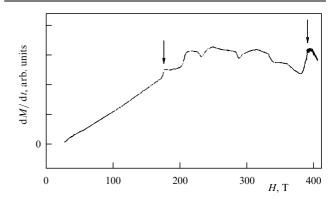


**Figure 4.** Theoretical magnetization curves of an  $Mn_{12}Ac$  cluster calculated at  $J_1 = -150 \text{ cm}^{-1}$ ,  $J_2 = -60 \text{ cm}^{-1}$ ,  $J_3 = 60 \text{ cm}^{-1}$ ,  $J_4 = -30 \text{ cm}^{-1}$ :  $I, \tau = 0; 2, \tau = 0.49 (105 \text{ K});$  and  $J_3, \tau = 1.47 (316 \text{ K}).$ 

#### 4.2. Experiment

Magnetization measurements in  $Mn_{12}Ac$  were conducted in pulsed magnetic fields up to 750 T produced by a magneto-cumulative generator MC-1. The experimental procedure is described in [13]. The results of these experiments show that a change from the ferrimagnetic to the ferromagnetic state begins in fields of about 400 T and ends in fields of about 750 T. At low temperatures ( $\sim 2-4$  K), this magnetization process is quantum in character, that is, much the same as presented in Fig. 4. It should be noted that in the region of magnetic fields from 150 to 400 T, a sequence of jumps is

observed whose nature is presently unclear (Fig. 5). To clarify the nature of these jumps, additional investigation is required.



**Figure 5.** Experimental field dependence of the time derivative of magnetization in  $Mn_{12}Ac$  at T=4.2 K.

Closing this section, we call attention to a further interesting and important property of high-spin clusters. The case in point is quantum magnetic hysteresis. This new phenomenon discovered and studied in recent years [27–31], as applied to the Mn<sub>12</sub> molecule, has attracted considerable attention of experts in the field of information technology. In this case, bistability and magnetic hysteresis manifest actually themselves in a single molecule, that is, the volume required for writing a bit of information is equal to that occupied by a single molecule. Such molecules realize Feynman's idea put forward more than 30 years ago that it is in principle possible to write information in a single molecule. Another important property of these high-spin molecules intimately related with this is 'microscopic quantum tunneling of magnetization'. This property is of interest from the viewpoint of realization of the 'quantum calculation' concept (see in detail, for example, [32, 33] and the literature cited there). In all these effects, a molecule manifests itself as an object with a specified spin which is the vector sum of the spins of all Mn ions making up the molecule.

The investigation of the magnetic properties of such a molecule in megagauss magnetic fields gives information on all possible spin states of the molecule beginning with the lowest-spin state S=10, when the spins of the Mn<sup>3+</sup> ions are aligned antiparallel to that of the Mn<sup>4+</sup> ions, and ending with the highest-spin state S=22, when these spins are aligned parallel to each other. This allows one to obtain information on exchange interactions between spins in this molecule, which is of importance for creating new nanoclusters with specified magnetic properties.

#### 5. Conclusion

In summary, one can state with assurance that measurements of the magnetic properties of materials in ultra-high magnetic fields is of considerable promise as a tool for studying molecular magnetism.

In particular,

(1) ultra-high magnetic fields allow one to obtain complete information on the properties of magnetization processes in many physically important systems such as metamagnets, f-d magnets with heavy rare-earth metals, atomic and nanometer-scale magnetic clusters [34];

- (2) atomic and nanometer-scale magnetic clusters clearly show quantum behavior in magnetization processes in contrast to most micro- and macrostructured materials;
- (3) jump-type anomalies of magnetic and magnetooptical effects caused by field-induced phase transitions open new possibilities for direct measurements of exchange and spin orbit coupling constants in solids, clusters, high-spin molecules [25], and ions [26].

Taking into account the special importance of studying the properties of magnetic nanoclusters, it should be noted that in  $Mn_{12}Ac$  clusters

- (4) phase transitions with retaining ferrimagnetic properties are realized in a field range from 180 to 750 T;
- (5) at low temperatures, the magnetization process is clearly quantum in nature;
- (6) the values of exchange constants commonly used in the literature are grossly overestimated.

#### References

- Pavlovskii A I, Kolokol'chikov N P, Tatsenko O M, in Megagauss Physics and Technology (Ed. P J Turchi) (New York: Plenum Press, 1980)
- 2. Goto T et al. Solid State Commun. 72 945 (1989)
- 3. Sakakigura E et al. J. Phys.: Condens. Matter 3 3381 (1991)
- 4. Wohlfarth E P, Rhodes P Philos. Mag. 7 1817 (1962)
- Levitin R Z, Markosyan A S Usp. Fiz. Nauk 155 623 (1988) [Sov. Phys. Usp. 31 730 (1988)]
- Dubenko I S et al Zh. Eksp. Teor. Fiz. 107 548 (1995) [JETP 80 296 (1995)]
- Dubenko I S, Kolmakova N P, Levitin R Z J. Magn. Magn. Mater. 153 207 (1996)
- Zvezdin A K Pis'ma Zh. Eksp. Teor. Fiz. 58 744 (1993) [JETP Lett. 58 719 (1993)]
- Evangelista L R, Zvezdin A K J. Magn. Magn. Mater. 140 144 1569 (1995)
- Dubenko I S et al. Pis'ma Zh. Eksp. Teor. Fiz. 64 188 (1996) [JETP Lett. 64 202 (1996)]
- Duc N H, Hien T D, Givord D J. Magn. Magn. Mater. 104-107 1344 (1992)
- 12. Brommer P E et al. Physica B 183 364 (1993)
- 13. Bykov A I et al. *Physica B* **216** 215 (1996)
- 14. Minkiewicz V J, Fujii Y, Yamada Y J. Phys. Soc. Jpn. 28 443 (1970)
- 15. Beckman O, Knox K Phys. Rev. 121 376 (1961)
- 16. Heeger A J, Beckman O, Portis A M Phys. Rev. 123 1652 (1961)
- Vedernikov N F et al. Zh. Eksp. Teor. Fiz. 93 2161 (1987) [Sov. Phys. JETP 66 1233 (1987)]
- 18. Muhin A A et al. Physica B (1998) (to be published)
- 19. Gatteschi D et al. Science 265 1054 (1994)
- 20. Sessoli R et al. Nature (London) 356 141 (1993)
- 21. Kahn O Comments Condens. Matter Phys. 17 (1) 39 (1994)
- 22. Papaefthymiou G C Phys. Rev. B 46 10366 (1992)
- Mukhin A A, Saĭko G V, Zvezdin A K "Molekulyarnye magnitnye materialy" (Preprint IOF RAN No 9 (Moscow: Institute of General Physics, 1994)
- Zvezdin A K, Popov A I Zh. Eksp. Teor. Fiz. 109 2115 (1996) [JETP 82 1140 (1996)]
- 25. Sessoli R et al J. Am. Chem. Soc. 115 1804 (1993)
- Zvezdin A K et al. Zh. Eksp. Teor. Fiz. 109 1742 (1996) [JETP 82 939 (1996)]
- 27. Sessoli R et al. *Nature* (London) **365** 141 (1993)
- 28. Friedman J R et al. Phys. Rev. Lett. 76 3830 (1996)
- 29. Thomas L et al. Nature (London) 383 156 (1996)
- 30. Dobrovitskii V V, Zvezdin A K Europhys. Lett. 38 377 (1997)
- 31. Gunter L Europhys. Lett. 39 1 (1997)
- Dobrovitskiĭ V V, Zvezdin A K, Popkov A F Usp. Fiz. Nauk 166 439 (1996) [Phys. Usp. 39 407 (1996)]
- Dobrovitskii V V, Zvezdin A K, in Nonlinear Microwave Signal Processing (Eds R Marcelli, S A Nikitov) (Dordrecht, Boston: Kluwer Academic Publishers, 1996) p. 29

Zvezdin A K et al., in *Itinerant Electron Magnetism: Fluctuation Effects* (NATO ASI Series, Partnership Sub-Series 3, Vol. 55, Eds D Wagner, W Brauneck, A Solontsov) (Boston, Mass.: Kluwer Academic Publishers, 1998) p. 285