Discrete Energy Spectrum of Nonlinear Spin Ensembles in the Ferrimagnet K_{0.4}\{Cr(CN)_6\}\{Mn(R/S)-pn\}(R/S)-pnH_{0.6}

A. D. Talantsev, M. V. Kirman, and R. B. Morgunov*

Institute of Problems of Chemical Physics, Russian Academy of Sciences, pr. Akademika Semenova 1, Chernogolovka, Moscow oblast, 142432 Russia
* e-mail: morgunov2005@yandex.ru

Received May 26, 2016

Abstract—Series of spontaneous magnetization jumps have been observed on the background of a continuous relaxation of the magnetic moment of a molecular ferrimagnet in a constant magnetic field. A statistical analysis of the set of data has demonstrated the presence of at least two modes in the distribution of magnetization reversal jumps over their amplitude. It has been found that the number of modes in the distribution depends on the magnetic field and temperature and characterizes a discrete energy spectrum of nonlinear spin ensembles formed at low temperatures. The continuous component of the magnetic relaxation corresponds to the motion of domain-wall arrays in the temperature range from 20 to 50 K and to relaxation processes in the spin-soliton lattice at temperatures ranging from 2 to 10 K.

DOI: 10.1134/S1063783417010334

1. INTRODUCTION

In chiral molecular magnets, the combination of the magnetic anisotropy of the sublattices, the asymmetric and symmetric exchange interactions, as well as a large lattice period (greater than 10 Å), lead to the formation of unusual nonlinear spin structures (spin ensembles), the response of which is observed in anomalous temperature dependences of the magnetization [1], the generation of nonlinear spin excitations by a microwave magnetic field [2], the bistability of a ferromagnetic resonance [3], and the Peierls dynamics of domain walls [4]. For thin films of gel-magnets (MnSi, FeGe, (FeCo)Si), one of the manifestations of the formation of a nonlinear magnetic phase (conical, helical, or lattice of skyrmions) is a jump-like change of the magnetization in a magnetic field after it reaches a certain critical value [5–7].

The magnetization jumps can have a stochastic nature. The most common types of nonlinearity in inorganic magnets are Barkhausen jumps that occur in the case of an irreversible displacement of the domain walls due to the decay of a single-domain state or as a result of the nucleation of new domains [8]. Of course, the magnetization reversal and jumps of the magnetization due to the motion of domain walls are also not excluded in molecular magnets, however, almost nothing is known about domains in these types of materials. In molecular magnets, chaotic jumps of the magnetization can be caused by the rearrangement of the spins into ferromagnetic and antiferromagnetic segments of linear chains of atoms, which form the molecular crystal [9]. Recent papers [10, 11] reported on the observation of stochastic jumps of the demagnetization in a chiral molecular magnet, where they correspond to nonlinear transformations of the internal structure of the spin solitons. Examples of the possible theoretical simulation of these events are presented in [12]. Similar experimental results were obtained more recently for the chiral gel magnet Cr_{1/3}NbS_{2}, in which the discreteness of the change in the magnetization was caused by the formation of spin-soliton lattices [13]. However, from the results of the aforementioned studies, it was impossible to judge both the sizes of ensembles of particles with spins and the discrete energy levels and their dependence on the magnetic field and temperature.

The purpose of this study was to determine statistical regularities of the jump-like magnetization reversal of the molecular ferrimagnet K_{0.4}\{Cr(CN)_6\}\{Mn(R/S)-pn\}(R/S)-pnH_{0.6}, as well as to verify the hypothesis about the presence of a discrete energy spectrum of nonlinear spin ensembles (solitons) in this material.

2. SAMPLES AND EXPERIMENTAL TECHNIQUE

The chemical synthesis and X-ray diffraction analysis of the K_{0.4}\{Cr(CN)_6\}\{Mn(R/S)-pn\}(R/S)-pnH_{0.6} single crystals, as well as the rapid testing of their magnetic properties (coercive force, Curie temperature), were described previously in [14]. A single crystal had
the form of a needle-like plate $\approx 1.5 \times 0.5 \times 0.1$ mm in size.

The magnetic moment $M$ of the samples was measured on a Quantum Design MPMS 5XL SQUID magnetometer in a constant magnetic field. The absolute error in the measurements of the magnetic moment was equal to $\approx 10^{-7}$ emu. The measurements were performed in the range of temperatures $T = 2$–$50$ K, at which the sample was in a magneto-ordered ferrimagnetic state (the Curie temperature was 53 K). During the measurement, the temperature of the sample was maintained constant with an accuracy of 0.1 K. Before the measurements, the sample was cooled in a zero magnetic field and then magnetized to saturation in a constant magnetic field $H_{\text{sat}} = 400$ Oe for a time $t_0 = 5$ min (see Fig. 1a and the schematic dependence in the upper part of Fig. 2a).

During the measurement of the demagnetization curves $M(H)$, the magnetic field was decreased in steps from $+28$ to $-4$ Oe with a switching step $\Delta H = 2.0$–$0.2$ Oe (Fig. 1a). In a series of experiments on the measurement of the relaxation of the magnetic moment $M(t)$, the sample was placed in a constant demagnetizing field $H$ for a time of 25 min (Fig. 2a). The time dependences of the magnetic moment $M(t)$ were obtained for the range of magnetic fields $H$ from $+30$ to $-50$ Oe.

3. RESULTS AND DISCUSSION

3.1. Magnetization Jumps in an Alternating Magnetic Field

The field dependences of the magnetic moment $M(H)$ were measured in a magnetic field that was decreased from the saturation field $H_{\text{sat}} = 400$ Oe with a step $\Delta H = 2.0$–$0.2$ Oe at a temperatures below the Curie temperature $T < 53$ K (Fig. 1). At $T = 2$ K, the magnetic moment of the ferrimagnetic sample in saturation was equal to $M_{\text{sat}} = 2\mu_B$ (where $\mu_B$ is the Bohr magneton). This value corresponds to an antiparallel orientation of the spins of the Mn$^{2+}$ ions and Cr$^{3+}$ ions. The magnetic field was applied along the easy axis of magnetization. It was revealed that the demagnetization curves $M(H)$ of the chiral molecular magnet are characterized by stochastic jumps of the magnetic moment. These jumps occur when the magnetic field reaches the critical value $H_C$ (inset in Fig. 1b). The magnetization jumps are reproduced on each curve $M(H)$ at a temperature in the range from 2 to 50 K. With a decrease on the step of switching of the magnetic field from 2.0 to 0.2 Oe (accordingly, with a decrease in the magnetic field sweep rate), it was found that there is a series of jumps of the magnetic moment. The maximum number of jumps was observed when the magnetic field sweep rate had the minimum value of 0.00125 Oe/s.

With an increase in the temperature $T > 8$ K, the average magnetic field of the jump with the maximum amplitude $H_{\text{cm}}$ is shifted toward weak magnetic fields (Fig. 1b). The measurement of the demagnetization curves $M(H)$ at low temperatures $T < 8$ K with a temperature step $\Delta T = 1$ K demonstrated that the dependence $H_{\text{cm}}(T)$ is nonmonotonic: at $T \approx 8$ K, there is a maximum (Fig. 1b).

In order to obtain the statistical distribution of the jumps of the magnetic moment and to determine the

![Fig. 1.](image-url)