

# Unusual magnetic phenomena in dynamic torsion studies of fullerene $\text{Rb}_3\text{C}_{60}$

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In dynamic experiments using a highly sensitive torsion technique, the magnetic properties of the doped  $\text{Rb}_3\text{C}_{60}$  single crystal are studied over a wide temperature range. The critical temperature of the superconducting transition  $T_c$ , the critical magnetic field  $H_{c1}$ , the pinning forces of both the Abrikosov vortices and the magnetic moments of dipoles at  $T \gg T_c$ , at which relaxation processes are important, are determined and investigated. It was shown that the structural transition  $Fm\bar{3}m-Pa\bar{3}$  at  $T = 260$  K is accompanied by a reformation of the magnetic structure. Both the usual peaks at  $T \sim 160$ – $250$  K and the not quite ordinary peaks of gigantic absorption of the energy of the oscillations, which shift to room temperature over time, were observed. The phenomenon is discussed in the framework of the cooperative Jahn–Teller effect, taking into account possible orbital and spin ordering. The relaxation processes at room temperatures ( $T \sim 300$  K) were studied. After many days of exposure (annealing) of fullerene at  $T \geq 40$  °C, in experiments with cooling from room temperatures, phenomena typical for a superconducting transition with giant pinning and damped oscillations were observed at  $T \sim 200$  K. The results of studies of magnetic phase transitions at  $T = 180$ – $200$  K in pure fullerite  $\text{C}_{60}$  (99.98% Term. Sublimed), which are related to the detected unusual magnetic phenomena in fullerene  $\text{Rb}_3\text{C}_{60}$ , are presented.

Keywords: fullerite,  $\text{Rb}_3\text{C}_{60}$ , Jahn–Teller effect, torsion vibration technique, trapped magnetic flux, molecular rotators.

## 1. Introduction

The discovery of the unique fullerene structures began with the predictions in 1970 by Osawa and Yoshida [1] and the quantum chemical calculations of Bochvar and Halperin [2] and only in 1985 did Kroto and his colleagues manage to synthesize the  $\text{C}_{60}$  molecule [3]. An intensive search began for ways to use fullerenes in electronics, biology, medicine and other fields. Having discovered the “bridge” between inorganic and organic matter, large-scale studies (including magnetic characteristics) of fullerenes and fullerides were started in scientific laboratories. The magnetic properties of fullerene superconductors are discussed in a rather extensive review [4].

Despite the relative simplicity of the  $\text{A}_3\text{C}_{60}$  structures ( $\text{A} = \text{K}, \text{Rb}$ ), the physical processes that determine the characteristics of this system, especially at low temperatures, have not been fully studied. So, for example, it still remains unclear what the role of the electronic, structural, and dynamic properties of such systems is in the relatively high

values of  $T_c$  — the temperature of the transition to the superconducting state. The nature of magnetic phase transitions in fullerenes at  $T > T_c$  also remains unclear. It is known that at temperatures close to room temperature,  $\text{A}_3\text{C}_{60}$  has a face-centered cubic (fcc) lattice, in which two alkali metal ions in tetrahedral (T) cavities fall on one  $\text{C}_{60}$  molecule and one in the octahedral (O) cavity of the crystal lattice. Sizes containing T and O cavities (T and O state of ions), according to NMR measurements, are correlated as 2:1. In this phase,  $\text{C}_{60}$  molecules rotate freely with a reorientation time of  $10^{-11}$  s. With decreasing temperature, a structural transition  $Fm\bar{3}m-Pa\bar{3}$  occurs [5–8] — to a phase having a simple cubic lattice, which is characterized by partial orientational ordering of the axes of rotation of  $\text{C}_{60}$  molecules along the spatial diagonals of the  $\langle 111 \rangle$  cube. In crystalline pure  $\text{C}_{60}$ , this transition was observed at  $T = 260$  K. In fullerenes doped with alkali metals, the temperature of this structural transition has not been precisely determined, although it is assumed that it occurs at higher  $T$ , since the presence of the alkali metal “interferes” with the free rotation of  $\text{C}_{60}$  mole-

cules. In NMR experiments on a  $K_3C_{60}$  sample [9,10], the peak associated with the structural transition overlaps with another, wider peak, presumably due to the appearance of the  $T^1$  state of the alkaline ion.

At high temperatures, all T states are equivalent. An additional resonant peak was observed in [11] during NMR studies of  $^{87}Rb$  below 370 K. It has been suggested that this peak is associated with a modified ( $T^1$ ) state of one of the tetrahedral (T)  $Rb^+$  states. Similar results and an additional peak were observed for  $K_3C_{60}$  [9,12] at  $T \sim 200$  K in studies of spin-lattice relaxation. In [9], this peak was interpreted as a phase transition in the system at these temperatures. Several mechanisms of this phenomenon have been proposed, in particular, ordering of the axes of rotation of  $C_{60}$  molecules, Jahn–Teller  $C_{60}^{3+}$  distortion,  $Rb^+$  clustering or modulation of charge carrier density, and also clarification for  $T-T^1$  cleavage of NMR lines by alkali cation vacancies. Thus, while there is no unambiguous explanation of the nature of this phenomenon (effect).

### *1.1. Methodology and superconducting characteristics*

As far as  $A_3C_{60}$  samples change (lose) their characteristics in air, including their superconducting ones they were sealed (placed) in quartz or glass ampoules (capsules). Therefore, for their study, the use of other direct methods, such as x-ray or transport measurements is difficult, because the sensitivity of the x-ray method decreases and the supply of electrical contacts is excluded. The SQUID magnetometer, which is very sensitive to the presence of magnetic moments in the samples under study, remains the most common measurement technique. However, in SQUID experiments, in contrast to torsion (vibrational dynamic) ones, the samples are fixed (motionless) in capsules. The latter does not allow one to detect (observe) such unusual phenomena as the interaction between fixed (pinned) and unpinned magnetic moments and associated dissipative processes, as well as relaxation phenomena and changes (reformation) of the magnetic structure relative to the applied external magnetic field with changing temperature.

In this work, to study the magnetic properties of  $Rb_3C_{60}$  fullerene, we used the torsion vibration method implemented using an automated multidisciplinary setup [13] with sensitivity at the level of a SQUID magnetometer [14]. The studies were carried out at low-frequency (0.1–1 Hz) oscillations in a magnetic field with a strength  $H$  that is constant and transverse to the axis of rotation of the sample and showed a significant influence on the results of the experiment history, the value of  $H$ , the initial orientation of the sample, and also the direction of change sample temperature (cooling or heating).

The torsion technique used is particularly sensitive to the reorientation of the magnetic moments of the materials under study in external magnetic fields. Since each  $C_{60}$  molecule can be represented as a magnetic dipole, such experiments will be informative of the reorientation of fullerene

molecules in the normal state at  $T > T_c$ . It can be assumed that this method will detect not only the structural transition  $Fm3m-Pa3$  [5–8], but also the transition associated with the appearance of the  $T^1$  state in  $Rb_3C_{60}$ .

The torsion oscillation method was first used to study the energy loss (dissipation) in the mixed state of superconductors in [15,16], which showed a rather high sensitivity ( $10^{-17}$  W) of the torsion system. Subsequently, this method was applied to determine the critical parameters of hard superconductors, such as  $T_c$  or the first critical field  $H_{c1}$  [17–20], to study the anisotropy of the pinning force  $F_p$  in high-temperature oxide superconductors (HTSC) [21], as well as “congenital” (intrinsic) [13,22–25] characteristics of HTSC samples. In addition, by studying dissipative processes near the temperature of superconducting transition  $T_c$ , one can observe and study the effects of the “melting” [26,27] of the Abrikosov magnetic vortex lattice [28], both in high-temperature superconductors and in fullerenes.

In this work, as in [14], we investigated the superconducting and magnetic properties of  $Rb_3C_{60}$  fullerene at temperatures from 4.2 K up to  $T = 290$  K, as well as magnetic phases in the normal state at  $T > T_c$  in temperature ranges from 80 to 300 K as the sample is heated in one case, and in another, from room temperature to 150 K as it cools. A sample located in a quartz capsule and suspended on a thin thread (with known elastic characteristics) oscillated in an external constant magnetic field  $\mathbf{H}$  directed perpendicular to the axis of rotation of the sample, the temperature of which slowly increased or decreased. The temperature dependences of the frequency  $\omega$  and the damping decrement of the oscillations  $\delta$ , of the oscillating sample in the programmed amplitude ranges ( $\sim 1^\circ$ ) were measured.

In the absence of magnetic moments in the sample, the dissipation and vibration frequency are independent of the external magnetic field. For example, this is observed for:

- i) a superconducting sample oscillating in magnetic fields  $H < H_{c1}$ , or
- ii) when the internal moments are zero or disoriented and not fixed.

The presence of the pinned Abrikosov vortices and (or) magnetic dipoles creates a nonzero magnetic moment  $\mathbf{M}$  in the sample. The interaction between  $\mathbf{M}$  and  $\mathbf{H}$  creates a moment  $\boldsymbol{\tau} = \mathbf{M} \times \mathbf{H} \sin \alpha$ , where  $\alpha$  is the angle between  $\mathbf{M}$  and  $\mathbf{H}$ . This additional moment  $\boldsymbol{\tau}$  acts on the oscillating system, as a result of which the dissipation  $\delta$  of the vibrational energy and the vibrational frequency  $\omega$  depend on the external magnetic field.

As shown in [29], for mixed state superconductors, the interaction between the pinned and non-pinned (free) vortices plays an important role in dynamic-vibrational processes. It is known that the pinning force also substantially depends on temperature, for example, it tends to zero as it approaches  $T_c$ . In this case, the concentration of free vortices increases and the value of the oscillation period  $t$  sharply increases (the frequency  $\omega$  decreases). In our experiments,

as the temperature of the  $\text{Rb}_3\text{C}_{60}$  fullerene studied in the normal state increased or decreased, not only did the relative concentration of pinning and free magnetic dipoles change, but also their orientation with respect to the constant external magnetic field  $\mathbf{H}$ , which was recorded by the angle of rotation of the sample accurate to  $10^{-4}$  rad. Undoubtedly, when approaching the temperature at which a structural and (or) magnetic phase transition occurs, it will be fixed in a “simple” or sometimes not quite ordinary, even gigantic (as will be shown later) change in the frequency and damping of oscillations.

The object of research was a sample of  $\text{Rb}_3\text{C}_{60}$ , consisting of single-crystal  $\text{C}_{60}$  doped with Rb by evaporation. The details of the sample preparation procedure and its corresponding characteristics were described in detail in [30,31], where the temperature dependence of the real ( $m'$ ) and imaginary ( $m''$ ) parts of the ac susceptibility, measured by a SQUID magnetometer, are shown in Fig. 1. A sharp drop in the magnetic susceptibility is due to the transition to the superconducting state at  $T_c = 30.9$  K. The sharp peak in the  $m''(T)$  dependence close to  $T_c$  indicates that the sample is in the superconducting state, although several other peaks are also visible at lower temperatures. These peaks are explained by dissipation due to weak links. Such granularity appears in the sample due to the presence of nonsuperconducting impurities (undoped  $\text{C}_{60}$ , or, most likely,  $\text{Rb}_{x \neq 3}\text{C}_{60}$ ) or due to the block structure of orientationally ordered regions of the  $\text{C}_{60}$  crystal. Using the method proposed by Angadi [32] and later developed for fullerene superconductors [33], the grain size was estimated as  $R \approx 350$   $\mu\text{m}$ . This a very large sample (with dimensions of approximately  $3.3 \times 2.7 \times 1.3$  mm) has several noticeable large grains with sizes up to several tenths of a millimeter, or possibly extensive granular regions. However, each grain can be considered as an  $\text{Rb}_3\text{C}_{60}$  single crystal with a good crystalline structure.

For comparison with the results (static) obtained by the SQUID magnetometer, we present the results of dynamic experiments on the same  $\text{Rb}_3\text{C}_{60}$  sample using the axial-torsional vibration method. Figure 2 shows the dependences of the period  $t = 2\pi/\omega$  and the damping decrement  $\delta$  with increasing temperature from  $T = 4.2$  K, after the FC (field

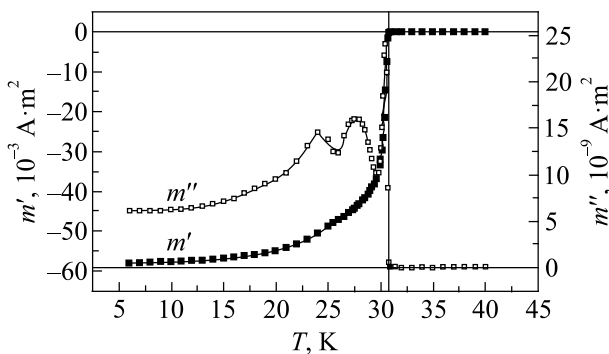


Fig. 1. Temperature dependences of magnetic susceptibility of fullerene  $\text{Rb}_3\text{C}_{60}$ , measured by SQUID magnetometer [30,31].

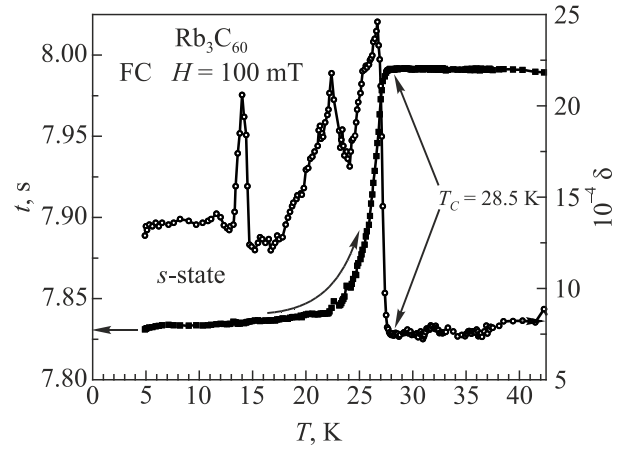


Fig. 2. Temperature dependences of the period  $t$  and decrement of oscillations damping  $\delta$  for  $\text{Rb}_3\text{C}_{60}$  after the FC procedure,  $H = 100$  mT [36,37].

cool) procedure in a magnetic field  $H = 100$  mT, i.e., transferring the sample to the superconducting state ( $s$ -state) and measurements in the same magnetic field. Such a procedure makes it possible to “freeze” (fix) both the vortex flow and magnetic dipoles (ordered state) along the  $\mathbf{H}$  direction and, as the sample is heated and approaches  $T = T_c$ , observe phenomena associated with reorientation of the system of magnetic moments and transition to a disordered state.

Obviously, such a transition will be accompanied by an increase in the damping of the oscillations up to the critical temperature, and then by a sharp decrease in the damping at  $T = T_c$ , when the vortex filaments “disappear”. When comparing the results obtained in statics (Fig. 1) with dynamic ones (Fig. 2), the similarity of the nature of the dependences draws attention. Three peaks in the imaginary part of the susceptibility  $m''$  are comparable with three peaks in the temperature dependence of the damping decrement  $\delta$ , but in the latter the first two peaks are shifted to low temperatures. The critical temperature determined from the real part of the ac susceptibility  $m'$  is  $T_c = 30.9$  K, while the temperature determined based on the temperature dependences of the period and decrement of vibration damping is  $T_c = 28.5$  K (Fig. 2). This discrepancy, presumably, is caused by an error in determining the temperature of the sample due to the inability to stick the thermometer and lead wires to the freely axially oscillating sample capsule. The temperature was measured at the sample level in the immediate vicinity of the oscillating system.

In the superconducting state ( $s$ -state), changes in both the frequency and the damping decrement can be explained by the motion at oscillations of the pinned vortices relatively free ones, as well as their interaction with the magnetic moments of the dipoles. This rather complex process at  $T > T_c$  ( $n$ -state) is somewhat simplified when, with a change in temperature, all the usual and not quite ordinary observed phenomena can be attributed only to magnetic phase transitions and (or) reorientations relative to the di-

rection  $\mathbf{H}$  of the magnetic moments of individual dipoles. Such an argument is valid only under the condition that there are no separate superconducting regions (grains) with Abrikosov vortices frozen in them.

In this work, we focus on the dynamics of these processes in the seemingly “normal” state ( $n$ -state) at temperatures above the lowest-temperature critical temperature  $T_{c1} = 28.5$  K of  $\text{Rb}_3\text{C}_{60}$  single-crystal. Figure 3 shows the temperature dependence of the period  $t$  and the attenuation decrement  $\delta$  in the range from helium to room temperatures (from 5 to 290 K), after the FC procedure  $H = 100$  mT.

As mentioned above, such a procedure is able to “freeze” Abrikosov’s vortex filaments in superconducting “drop” regions, as well as individual single-crystal grains, which can differ in critical temperatures  $T_c$ . As the temperature rises, these areas become normal the  $n$ -state and release the pinned vortex filaments, the viscous movement of which along the sample matrix leads to an increase in the dissipation of vibrational energy and a corresponding change in the oscillation period. These two characteristics  $t(T)$  and  $\delta(T)$ , measured in parallel and complementing each other, inform about the presence or absence of magnetic vortex filaments in the test sample, thereby allowing us to judge the state of the sample.

As can be seen from Fig. 3, after the clearly expressed  $T_{c1} = 28.5$  K (see Fig. 2), the anomalies at 54.5 and 93 K are visible on the temperature scale, in our opinion, related to the features of the transition from the superconducting to the normal state. The observed anomalies are the attenuation peaks in the  $\delta(T)$  dependence at 130, 185 and 226 K, we believe are related and well correlated with the magnetic phase transitions that are observed in  $\text{C}_{60}$  fullerite and are described in Sec. 1.4. The decay peak at  $T = 260$  K is undoubtedly associated with the structural transition  $Fm3m-Pa3$ .

Attention is drawn to the slow decrease in the period  $t$  after  $T > T_{c1} = 28.5$  K up to  $T = 54.5$  K. We repeatedly observed this nature of the dependence in the study of multiphase HTSC Bi–Pb–Sr–Ca–Cu–O systems, while the

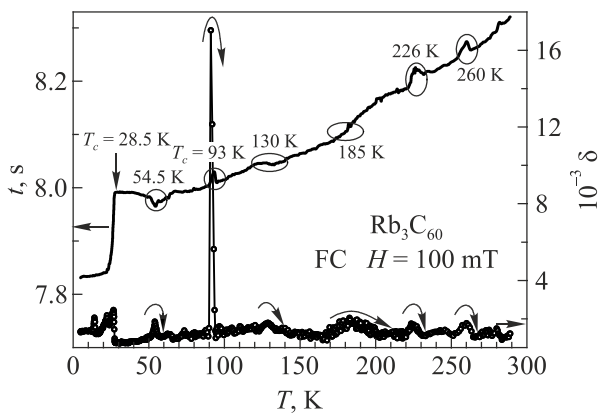


Fig. 3. Temperature dependences of the period  $t$  and damping decrement  $\delta$  with increasing temperature from  $T = 4.2$  to 290 K for  $\text{Rb}_3\text{C}_{60}$  (FC  $H = 100$  mT).

measurements of these  $t(T)$  dependences were duplicated by the standard four-contact method of measuring the electrical resistance  $R(T)$  and it was confirmed that superconducting transition takes place.

It can be assumed that the dependence  $t(T)$  shown for Fig. 3 for the  $\text{Rb}_3\text{C}_{60}$  fullerene after  $T > T_{c1} = 28.5$  K indicates the presence in the sample under study of at least a second and higher temperature phase with  $T_{c2} = 54.5$  K.

Superconductivity with a critical temperature  $T_c = 52$ – $54$  K in donor-doped fullerene  $\text{C}_{60}$  (K, Cs, Rb) was discovered back in [34]. According to investigations conducted by authors the metal state of the material and the superconductivity associated with it was the result of electron transfer from alkaline or alkaline earth ions to the  $\text{C}_{60}$  molecule, which in itself is an effective acceptor (which is why it is difficult to remove electrons from it). Comparing the obtained values with the well-studied dependence of  $T_c$  on the lattice parameters for electronically doped  $\text{C}_{60}$ , the authors suggest that in the future it is possible to achieve a  $T_c$  value significantly exceeding 100 K.

Figure 3 draws attention to the surge in the damping of the oscillations at  $T = 93$  K. This feature, by the nature of the dependences  $t(T)$  and  $\delta(T)$ , is very similar to the superconducting transition, but unfortunately, the absence of SQUID measurements by a magnetometer in this temperature range, and especially the electrical resistance  $R(T)$ , does not allow an unambiguous statement. It is possible that the features at  $T = 54.5$  and 93 K are associated with the manifestation of superconducting phases remaining in individual grains of the crystal. A fragment of this feature is shown in Fig. 4(a) on an enlarged scale (over the period) for  $\text{Rb}_3\text{C}_{60}$  fullerene, and also for comparison, Fig. 4(b) shows a typical dependence observed at critical temperatures  $T \approx T_c$  in multiphase bismuth cuprates after the FC procedure and, of course, related with the “captured” Abrikosov vortices. It can also be assumed that the features observed at  $T = 93$  K are related to the disinhibition of the rotational motion of  $\text{C}_{60}$  molecules upon transition from the orientation glass state to the orientationally disordered phase, which was observed in [35] during torsion studies of  $\text{C}_{60}$  fullerite in a magnetic field.

From our point of view, such an explanation of the observed phenomena is equally acceptable, both for the multiphase bismuth system and for the  $\text{Rb}_3\text{C}_{60}$  fullerene system (Fig. 4) containing individual rather large grains, which could after the FC procedure in the field  $H = 100$  mT to  $T = 4.2$  K “freeze” in itself and exhibit higher-temperature superconducting properties at temperatures  $T > T_{c1} = 28.5$  K.

It should be noted that in all studies of multiphase superconductors, the damping peak  $\delta(T)$  associated with Abrikosov vortices at temperatures  $T \approx T_c$  of one or another superconducting phase does not exceed  $\delta = 10^{-2}$ – $10^{-3}$ , while the maximum value of  $\delta$  falls at a temperature always lower than  $T_c$  (see Fig. 4), which is observed in these figures.



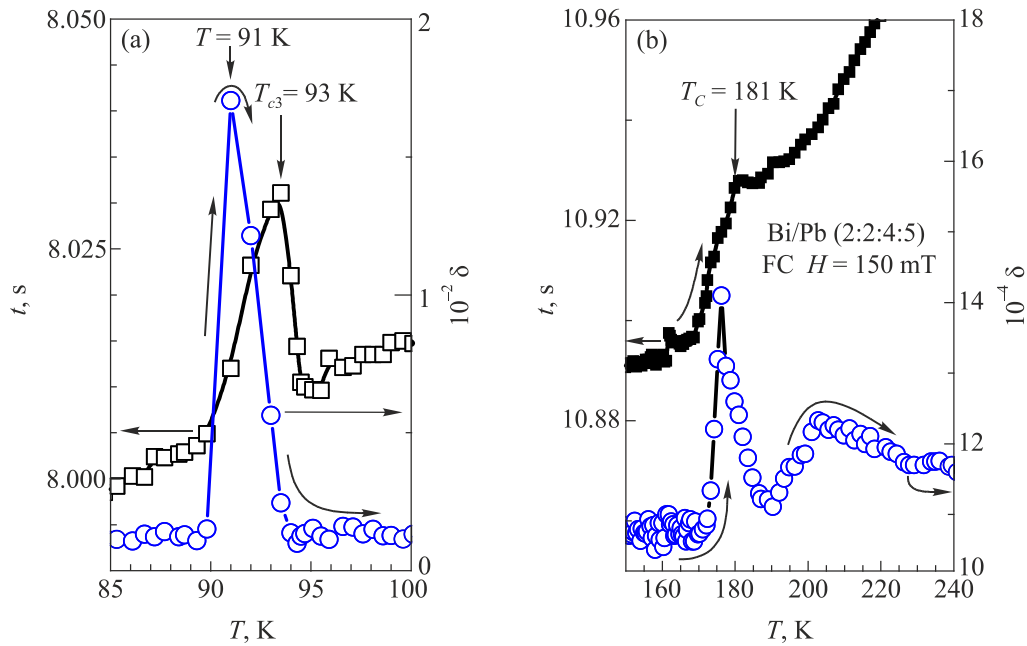


Fig. 4. Fragments of the temperature dependences of the period  $t$  and decrement  $\delta$  of vibrations damping for: (a) the  $\text{Rb}_3\text{C}_{60}$  fullerene on an enlarged (over the period) scale; (b) a multiphase bismuth cuprate  $\text{Bi/Pb (2:2:4:5)}$ .

Using the rotational mode [13] in torsion measurements, we determined at  $T = 4.2$  K the volumetric pinning force  $F_p \approx 20$  dyn/cm<sup>3</sup> [36,37] by the simple (non-contact) method proposed in [38,39], as well as the value  $H_{c1} = 15$  mT, which is in good agreement with the values obtained by other methods, see, for example, [4].

### 1.2. Magnetic properties in normal state of cooling fullerene $\text{Rb}_3\text{C}_{60}$ , “spontaneous” rotation and relaxation

The main experimental results of studying the temperature dependences of the oscillation period  $t$  in the normal state of  $\text{Rb}_3\text{C}_{60}$  fullerene obtained before annealing during slow (for 6.5 h) cooling from 280 to 197 K in a magnetic field  $H = 150$  mT are shown in Fig. 5. As can be seen, the process of changing (decreasing) the period  $t$  begins directly from room temperatures down to  $T \sim 260$  K — the temperature of the structural transition  $Fm3m\text{-}Pa3$  [5–8]. The gradual cooling of the sample and a decrease in the period of oscillations  $t$  from  $T \sim 222$  to 202 K probably indicates a process in the direction of magnetic ordering, when the total magnetic moment  $\mathbf{M} = \sum \mathbf{m}_i$  ( $\mathbf{m}_i$  is the magnetic moment of an individual dipole) is reoriented in the direction  $\mathbf{H}$ . If, at temperatures  $T = 225$  and 202 K (at minima of  $t$  values), a phase transition occurs even in individual grains, leading to a reformation and partial destruction of the existing relative magnetic order, then the latter leads to the fact that with further cooling, the oscillation period slightly increases (see Fig. 5). The peak in the temperature dependence of spin-lattice relaxation in  $\text{K}_3\text{C}_{60}$  at  $T \sim 200$  K can be explained [9,12] by a phase transition and several models for interpreting this phenomenon have been proposed, of which

(in our opinion) the Jahn–Teller distortion  $\text{C}_{60}^{3+}$  is most acceptable.

The transition shown in Fig. 5 at  $T \sim 200$  K is accompanied by an increase in the damping  $\delta(T)$  of the oscillations and, despite the spread in the values of  $\delta$ , a peak with the level  $10^{-2}$ – $10^{-3}$  is clearly seen. We note that it was in this temperature range at  $T \sim 200$  K, later on, after changing the experiment prehistory, the unusual phenomena of giant absorption of oscillations and pinning, which are described below in Secs. 1.3 and 1.4, were observed.

It is known that a magnetic field penetrates a superconducting sample if  $H > H_{c1}$  in the form of quantized Abrikosov vortices, which, by virtue of energy considera-

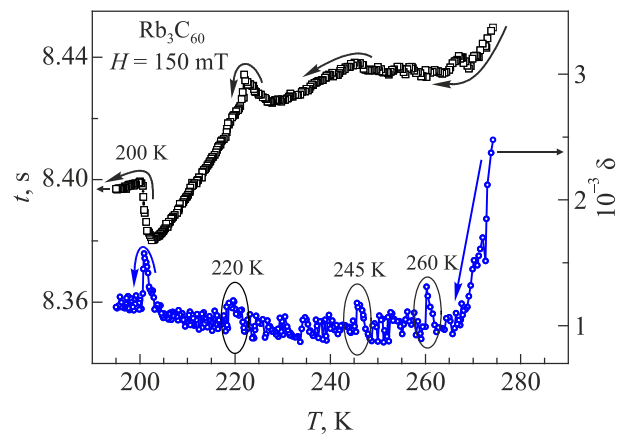


Fig. 5. Temperature dependences of the oscillation period  $t$  and the damping  $\delta$  of the oscillations obtained upon cooling of  $\text{Rb}_3\text{C}_{60}$  fullerene in a magnetic field  $H = 150$  mT before the magnetic-mechanical rotational annealing (MMRA) procedure (see Sec. 1.3).

tions, are pinned on existing structural defects. In the general case, the magnetic moment  $\mathbf{M}$  associated with these vortices is located relative to the external field  $\mathbf{H}$  at a certain angle  $\varphi$ , which is easily detected in the torsion method by “spontaneous” rotation of the sample. This rotation allows us to determine the value of the applied torque  $\tau = k\varphi$  ( $k$  is the torsional rigidity of the suspension thread).

Detailed studies of this phenomenon were carried out in [40] on a classical superconductor of the second kind Nb at  $T = 4.2$  K in a wide range of magnetic fields  $H_{c1} < H < H_{c2}$ . The anisotropy of the torque  $\tau$  was studied for various orientations of the sample  $\theta = 0-360^\circ$  (in increments of  $10^\circ$ ) with respect to  $\mathbf{H}$ , and a rosette figure was constructed, which showed the existence of certain orientations at which  $\tau = 0$ . Metallographic and electron microscopic analyzes revealed the corresponding crystallographic directions and showed that they are associated with the boundaries of single-crystal blocks, as well as with associated dislocations oriented along these directions. It was also shown [40] that the pinning force is maximum when the vortex structure is located along these directions.

HTSC samples exhibit anisotropic, moreover pronounced, properties, the magnetic properties of which are associated with the “intrinsic” [22] torque  $\tau = \mathbf{M} \times \mathbf{H} \sin \alpha$ . The latter is due to the strong difference in the effective electron masses along the  $c$  axis and in the basal  $ab$  plane ( $m_{ab}$ ).

These torques were experimentally observed, for example, in [23,24] on HTSC samples with  $c$ -oriented granules, as well as on samples textured by uniaxial compression [13,25].

However, let us return to the  $\text{Rb}_3\text{C}_{60}$  fullerene studied in dynamic (vibrational) experiments. As noted above, in the process of measuring the temperature dependences of the period  $t$  and the damping decrement  $\delta$ , a “spontaneous” rotation of the sample  $\varphi_0$  relative to the initial position  $\theta = 0$  in a magnetic field is recorded simultaneously with an accuracy of  $10^{-4}$  rad. This rotation can occur both clockwise and counterclockwise, depending on the initial orientation  $\theta$ . Moreover, in the process of “spontaneous” change of  $\varphi_0$ , before measuring the next point, the position of the measuring scale with photosensors is automatically adjusted so that the oscillations occur relative to  $\theta = 0$  on the scale of the device. The nature of the dependence  $\varphi_0(T)$  carries certain information about the processes of not only the reformation of the magnetic structure and reorientation of the magnetic moments of the dipoles with respect to  $\mathbf{H}$ , but also about the pinning force that rotates and keeps the sample rotated by one or another angle  $\varphi_0$ .

A rotation through the angle  $\varphi_0$  is the rotating torque  $\tau = k\varphi_0$  (in these experiments,  $k = 2.63 \cdot 10^{-4}$  N·m). Knowing the values of  $\tau$ , as shown in [41], one can also estimate the volumetric force of pinning

$$F_p = 0.75 \tau R^{-3} L^{-1}, \quad (1)$$

where  $R$  is the radius and  $L$  is the length of the sample. Estimation showed that  $F_p = 0,6$  dyn/cm<sup>3</sup> for the pinned

magnetic dipoles. Despite the small  $F_p$  values for magnetic dipole of fullerenes in the normal state, it can be recorded both in static and dynamic experiments. The value of  $F_p$ , as well as the nature of the dependences  $\varphi_0(T)$ , can vary significantly depending on the history of measurements, as well as on the initial orientation  $\theta$ . An increase in  $\varphi_0$  can be replaced by jump-like drops as the sample cools from room temperature.

It was noted that in most cases, upon completion of measurements and turning off the magnetic field, the oscillating sample, warmed up to room temperature and located in the residual field of the electromagnet  $H = 2$  mT, does not return to its original (zero) position and relaxes to it after many hours, and sometimes days. One example is shown in Fig. 6, when, after the next FC (field cool) experiment at  $H = 150$  mT, the sample heated from  $T = 77$  to 295 K even in the residual field of the electromagnet  $H = 2$  mT continues to relax for 2 h (pretty in an uneasy way) to its initial (at room temperature) values of the period  $t$  and the damping of oscillations  $\delta$ . It must be assumed that this process is caused by a continuing reformation in the system of magnetic moments of dipoles due to thermal fluctuations at  $T = 295$  K. It is enough to remove the magnetic field (remove the electromagnet from the system with the sample), as the sample returns to the position  $\theta = 0$  and the initial values of the frequency and damping of the oscillations are restored.

The magnetic moments of the  $\text{Rb}_3\text{C}_{60}$  fullerene dipoles are particularly sensitive to thermal fluctuations in the region of room temperatures  $T = (295 \pm 10)$  K. An increase in temperature even by several degrees relative to room temperature significantly changes the value and nature of the dependence of both the period of oscillations  $t$  and the angle of “spontaneous” rotation  $\varphi_0$ , which is shown in Fig. 7 in the field  $H = 150$  mT. The slow heating switched on at  $T = 295$  K when it reached  $T = 303$  K (after 24 min) was turned off because it was known that the melting point of the alkali metal (dopant) Rb  $T_m \approx 312$  K is only 10 degrees higher, which as the annealing temperature was in-

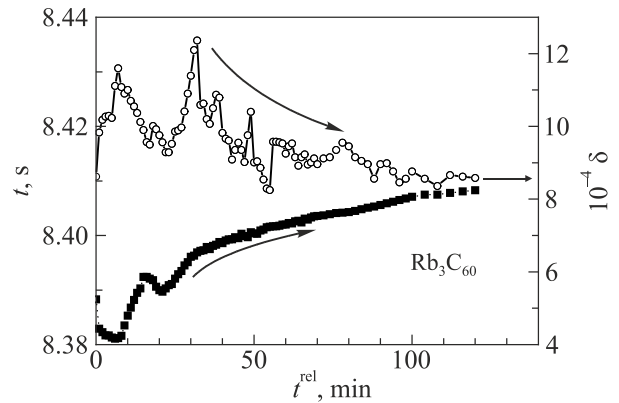


Fig. 6. Relaxation of the dependences of the period  $t$  and vibration damping  $\delta$  at  $T = 295$  K in the residual field  $H = 2$  mT, after FC measurements at  $H = 150$  mT.

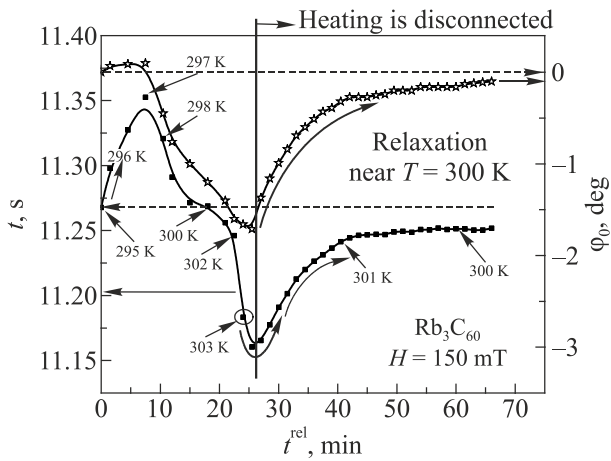


Fig. 7. Dependence of the oscillation period  $t$  and the angle of “spontaneous” rotation  $\varphi_0$  of the  $\text{Rb}_3\text{C}_{60}$  sample with heating and after  $T = 303$  K cooling in the field  $H = 150$  mT.

cluded in the program of subsequent experiments described in Sec. 1.4. After turning off the heating, the oscillating sample for almost an hour, relaxes to the initial orientation  $\theta = 0$ , the level of which is indicated by dashed lines. Extrapolation shows that the return will occur in no less than 15 h. A significant decrease in the oscillation period  $t$  (increase in frequency) at  $T = 303$  K indicates a facilitated process of reformation and reorientation of magnetic dipoles due to a decrease in the pinning forces with increasing temperature.

### 1.3. Giant absorption of oscillations

Before proceeding to the presentation of the not entirely ordinary results of measurements of the period  $t$  and the damping of the oscillations  $\delta$  performed for the  $\text{Rb}_3\text{C}_{60}$  sample in the normal state, it is necessary to describe the background history preceding these experiments, since namely it possibly clarifies the observed phenomena. The anisotropy of such quantities as  $\delta$  and  $t$  was studied at room temperature in a magnetic field  $H = 150$  mT at various initial orientations  $\theta = 0\text{--}360^\circ$  (36 points in increments of  $10^\circ$ ) and then back from  $360^\circ$  to  $\theta = 0$ , with the same step. Measurements lasted 20 days on average for 3 orientations per day. The transition from one orientation to another was carried out directly in a magnetic field  $H = 150$  mT. When measurements were continued the next day, the field was turned on at the last measured orientation and reorientation was performed, etc. In total, measurements were taken over 100 h. Such a lengthy procedure of magnetic-mechanical rotational annealing (MMRA) in the field  $H = 150$  mT, at  $T = 293$  K, led to unusual results on  $\text{Rb}_3\text{C}_{60}$  fullerene in subsequent studies.

Figure 8 shows the first observation of a giant absorption of oscillation energy after the MMRA procedure [36,37,42]. All experiments were carried out, under the same conditions, after a sharp cooling in a magnetic field

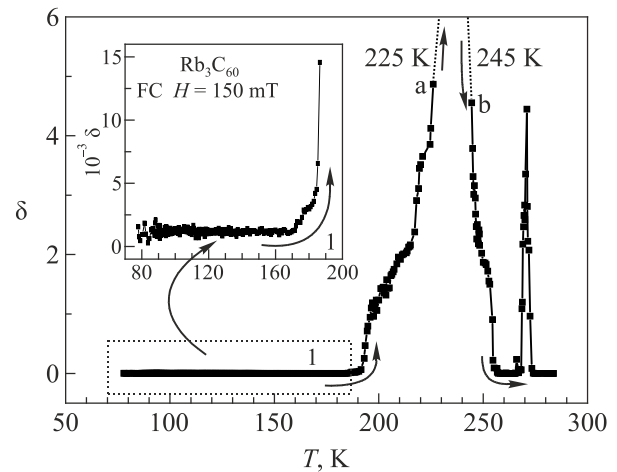
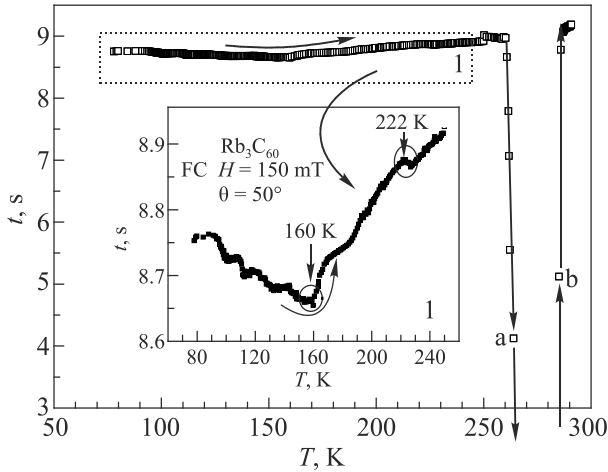


Fig. 8. Temperature dependence of giant oscillation damping  $\delta$  of  $\text{Rb}_3\text{C}_{60}$  fullerene after MMRA procedure at room temperature and FC in a magnetic field  $H = 150$  mT.

FC  $H = 150$  mT followed by a slow (for 8 h) heating from  $T = 80$  K to  $T \leq 300$  K.

From experiment to experiment (6 measurements per month), the attenuation peaks shifted to room temperatures. In subsequent multiple studies, it was noted that as the measurements were carried out, both the width and the height of the attenuation peaks sharply decrease without observing the region with giant dissipation. For example, the peak height at  $T = 205$  K became 2 orders of magnitude smaller than the original, and the width decreased down to 15 K. At the same time, the satellite peak at  $T = 271$  K shifted to room temperature  $T = 290$  K [36]. In the latest measurements, the peaks disappeared altogether, but special regions (kinks) at  $T = 255\text{--}260$  K began to appear on the  $t(T)$  dependences, which is apparently related to the structural transition  $Fm\bar{3}m\text{--}Pa\bar{3}$ .

Based on the observations made, it became obvious that in order to re-observe the giant absorption of oscillations with the initial level, it was necessary to change the measurement prehistory or to conduct again a sufficiently long magnetic “annealing”, because heating the sample to  $T \sim 300$  K for a short time did not produce significant changes. Sample  $\text{Rb}_3\text{C}_{60}$  was rotated relative to  $H$  by an angle  $\theta = 50^\circ$ . From this orientation (considering it to be zero), a “training” was conducted — MMRA for one day (and not 20 days as before) at  $T = 292$  K in the field  $H = 150$  mT. Only the oscillation period  $t$  was measured, depending on the orientation from  $\theta = 0$  to  $360^\circ$  and vice versa, in steps of  $10^\circ$ . The next day, in the FC experiment in the field  $H = 150$  mT, the dependences  $\delta(T)$  and  $t(T)$  again showed a gigantic oscillation damping but at higher temperatures. These results are shown in Fig. 9, which shows how a sharp decrease in the oscillation period  $t(T)$  begins at  $T \sim 260$  K from point (a) and returns from the unmeasured region to point (b) at  $T = 287$  K. In the inset of figure in the enlarged scale it is shown the interval  $T = 80\text{--}240$  K, with a typical “fracture” at  $T = 160$  K.

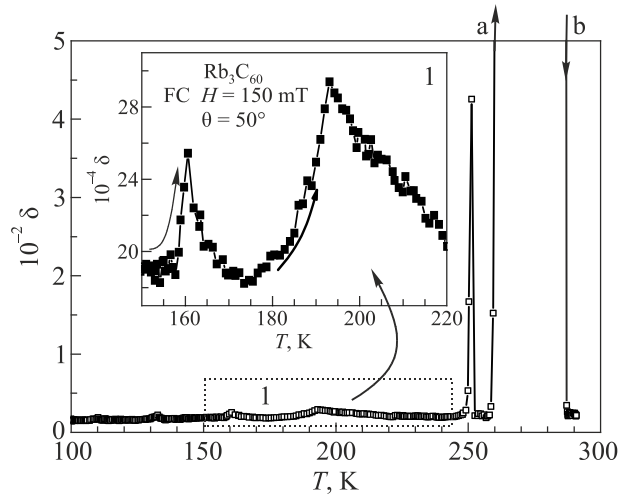


*Fig. 9.* Temperature dependence of the oscillation period  $t$  of fullerene  $\text{Rb}_3\text{C}_{60}$  in the field  $H = 150$  mT after reorientation to  $\theta = 50^\circ$  and magnetic “Workouts” — MMRA at room temperature.

Thus, after a reorientation to  $\theta = 50^\circ$  and the MMRA procedure, when the  $\text{Rb}_3\text{C}_{60}$  sample was heated, a sharp decrease in the oscillation period  $t(T)$  was observed (see Fig. 9), as well as an increase in the frequency with a subsequent decrease to the values at which our recording system does not work. We observed a similar behavior of the measured parameters when studying hard superconductors of the second kind, in which the pinning force of vortex filaments  $F_p$  was increased significantly after deformation by compression of niobium (Nb) single crystals. At small amplitudes and increased vibration frequencies, this phenomenon was called the “Hare Tail” effect by colleagues. We consider this comparison with the forces of pinning of Abrikosov’s vortices [28] to be appropriate since it rises question: we observe the “Giant absorption” of oscillations, and (or) the “Giant pinning”. In any case, after a successful buildup, we observed practically “undamped” oscillations visually at small unmeasured amplitudes. As the temperature rises, it gradually increases and both the amplitude and the oscillation period become measurable. At  $T = 287$  K, the sample returned to the position indicated by point (b) in Fig. 9, where the oscillation attenuation and the oscillation period began to be recorded by a programmed automatic system.

The corresponding dependence of the damping decrement  $\delta(T)$  is shown in Fig. 10, it can be seen that a change in the background, in particular, preliminary orientation at  $\theta = 50^\circ$  and “training” — magnetic annealing at  $T = 295$  K — led to the appearance of a giant absorption oscillation that is shifted to room temperature of the  $\text{Rb}_3\text{C}_{60}$  fullerene sample in the FC experiment in a constant magnetic field  $H = 150$  mT.

In early NMR experiments, some anomalies were observed in the temperature dependence of the spin-lattice relaxation times in this temperature range on  $\text{K}_3\text{C}_{60}$  [9,10] and  $\text{Na}_2\text{CsC}_{60}$  [43]. However, such anomalies were very weak and, as shown, their magnitude is close to the experi-



*Fig. 10.* Temperature dependence of oscillation attenuation  $\delta$  of fullerene  $\text{Rb}_3\text{C}_{60}$  in the field  $H = 150$  mT after reorientation to  $\theta = 50^\circ$  and magnetic “training” — MMRA at  $T = 295$  K. The inset shows in an enlarged scale the region  $T = 150$ – $220$  K.

mental error. In our studies, the magnitude of the effect is huge and manifests itself in torsion measurements, which are sensitive to the presence of free and oriented (pinned) magnetic moments of dipoles. The phenomena observed by us indicate that at these temperatures, a substantial reformation, reorganization in the magnetic structure of  $\text{Rb}_3\text{C}_{60}$  occurs.

It can be assumed that the number of pinned magnetic moments of dipoles decreases with increasing temperature, and the effect observed at  $T \sim 300$  K, after magneto-mechanical rotational annealing, is due to their reorientation. Although a superposition of several effects is not excluded, such as the reorientation of the magnetic moments of dipoles and the phase transition at  $T \sim 280$  K, or a first-order transition  $Fm3m$ – $Pa3$  associated with orientational ordering or disordering in a system of  $\text{C}_{60}$  molecules (for pure  $\text{C}_{60}$ ,  $T = 263$  K). In  $\text{Rb}_3\text{C}_{60}$ , this transition appears at a higher temperature due to the presence of Rb atoms in the cavities of the fullerite crystal lattice. A dynamic charge redistribution process is also possible in the  $\text{Rb}_3\text{C}_{60}$  system, which manifests itself at high temperatures and is associated with the appearance or decay of the modified tetragonal  $T^1$  fullerite state, which differs in the orientation of the five-membered  $\text{C}_{60}$  rings to  $\text{Rb}^+$  ions [11].

As for many orders of magnitude large in amplitude and wide in temperature peaks of damping oscillations, both in the region of  $T \sim 200$  K and higher (see Figs. 8–10), we do not have information about such a strong (giant) magnetic response in fullerene material in dynamic studies. Therefore, it can be assumed that it is not associated with a phase transition or dynamic overlap, but is caused by distortions of the lattice of  $\text{C}_{60}$  molecules and (or)  $\text{Rb}_3\text{C}_{60}$  lattice. We believe that the explanation for the giant magnetic response is associated with the manifestation of the Jahn–Teller effect (JTE), which can be confirmed by the experimental observations described below (Sec. 1.4).



#### 1.4. Giant pinning in $\text{Rb}_3\text{C}_{60}$ fullerene at $T \sim 200$ K after annealing

The above results of dynamic studies of the magnetic properties of  $\text{Rb}_3\text{C}_{60}$  fullerene showed a significant influence on them, both the experiment prehistory (FC, ZFC,  $H$  value), and the initial orientation of the sample in a magnetic field, as well as the procedure of MMRA in the field  $H = 150$  mT at  $T = 293$  K. Therefore, we will describe successively prehistory, possibly influencing the secondary stronger manifestation, that appears, then disappears and “walks” on the temperature scale of the effect of giant absorption of oscillations and (or) giant pinning. Note that a gigantic absorption of vibrational energy was observed in experiments when heating from 77 K to room temperatures only after FC and MMRA procedures [36,37,42].

On the other hand, even before carrying out these procedures, we noticed that heating of fullerene even by several degrees relative to room temperature significantly changes the characteristics under study (see Sec. 1.2, Fig. 7). It is known that the melting temperature of an alkali metal (dopant) Rb is  $T_m \approx 312$  K. We performed annealing for several days, keeping fullerene  $\text{Rb}_3\text{C}_{60}$  in the capsule at a temperature of  $T \geq 40$  °C, while the sample was outside the magnetic field, not counting the  $H \sim 0.5$  Oe field of the Earth.

Before proceeding to measurements with abrupt cooling of the sample in a magnetic field (FC), taking into account the procedures after which earlier (item 1.3, Figs. 8–10) unusual gigantic damping, as well as pinning of the magnetic moments of dipoles, were observed the temperature dependences of the decrement and the oscillation period of the annealed  $\text{Rb}_3\text{C}_{60}$  fullerene were measured as it slowly cooled from room temperature.

The results of the experiment in a magnetic field  $H = 150$  mT, which lasted for 4.5 h from  $T = 295$  to 212 K, are shown in Fig. 11. The period  $t$  of the fullerene  $\text{Rb}_3\text{C}_{60}$  oscillating in the programmed amplitude limits ( $A \sim 1^\circ$ ) at a critical temperature  $T = 211.5$  K sharply decreased and the effect of “the Hare Tail” appeared. Naturally, with a fall in  $t$ , the oscillation damping  $\delta$  also sharply increases and the sample goes into the temperature range inaccessible to measurements. Our attempts to rock the sample were unsuccessful during many hours of cooling to  $T = 150$  K and subsequent heating to  $T = 200$  K with the inclusion of a special heating stove in a cryostat under the sample. Unfortunately, the low power of the heater, designed for operation at helium temperatures, did not allow measurements at  $T > 200$  K, due to the coming equilibrium of heat supply and cooling from the stove and liquid nitrogen in the “jacket” of the cryostat, respectively.

In the process of cooling of fullerene from room temperature to  $T = 211.5$  K, the “spontaneous” rotation of the sample relative to the initial position was  $\varphi_0 = 5.6^\circ$ .

Moreover, during the first measurement, it jumps to this position abruptly at  $T \leq 211.5$  K (see Fig. 11), while in subsequent (repeated) measurements, jumps are not ob-

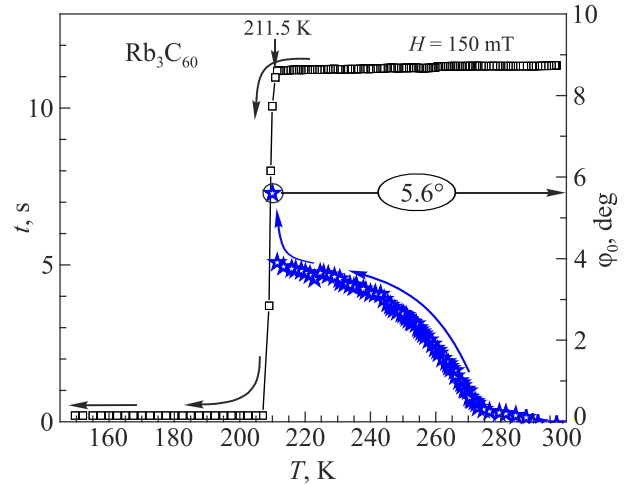


Fig. 11. Temperature dependences of the oscillation period  $t$  and the angle of “spontaneous” rotation  $\varphi_0$  of  $\text{Rb}_3\text{C}_{60}$  fullerene cooling from the room temperature in the field  $H = 150$  mT after annealing at  $T \geq 40$  °C.

served (see Fig. 14). The removal of the electromagnet from the sample restores normal vibrations and returns it to the position  $\varphi_0 = 0$ . The reverse input of the switched on electromagnet returns and locks the sample in position  $\varphi_0 = 5.6^\circ$ , in which it was located before the removal.

The definite (in this case, at  $\varphi_0 = 5.6^\circ$ ) torque  $\tau = k\varphi_0$  is 8.5911 dyn·cm, for which from relation (1) we obtain, for the pinning magnetic dipoles at  $T = 150$ –212 K, a rather large volumetric force pinning  $F_p = 2.15 \cdot 10^4$  dyn/cm<sup>3</sup>, which is comparable with the values obtained for Abrikosov vortex filaments in high-temperature superconductors (HTSC) at  $T < T_c$ .

The character of the dependence  $t(T)$  after annealing (Figs. 11 and 12) is very similar to the superconducting transition of this sample at  $T_{c1} = 28.5$  K, shown in Fig. 2 and in the inset in Fig. 12. The significant difference between these observations is that, at helium temperatures in

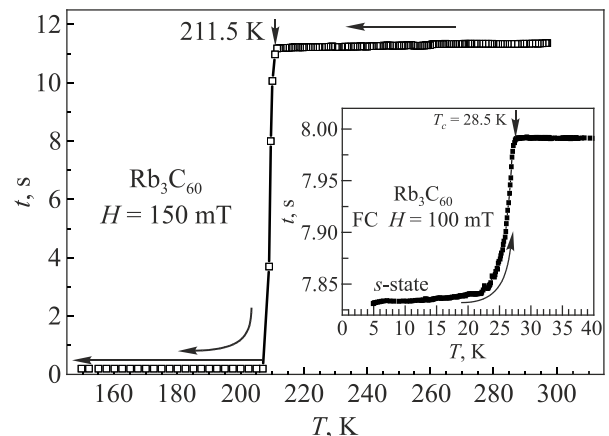


Fig. 12. Temperature dependences of the oscillation period  $t$  of fullerene  $\text{Rb}_3\text{C}_{60}$  in experiments at  $H = 150$  mT with cooling after annealing at  $T \geq 40$  °C. The inset (for comparison) shows the dependence  $t(T)$  with Fig. 2.

the superconducting state at  $T < T_c$  (see the inset in Fig. 12), the oscillation period was  $t \approx 7.8$  s, whereas after “annealing” at  $T \geq 40$  °C “Hare Tail” effect was observed — a sharp decrease in the oscillation period to  $t \approx 0.2$  s, which is  $\sim 40$  times less than the values after the true superconducting transition shown in the inset in Fig. 12. The magnitude of the period of oscillations in a magnetic field indicates a rather large force of pinning of Abrikosov vortices if  $T < T_c$  ( $s$ -state) or pinning of the magnetic moments of dipoles in the  $n$ -state at  $T > T_c$ . In any case, the result shown in Figs. 11 and 12 is the observation of “Giant pinning” on  $\text{Rb}_3\text{C}_{60}$  fullerene after “annealing”. It should be noted that a sharp decrease in the period of oscillations  $t$  below  $T = 211.5$  K is accompanied by the same sharp, but already increased, decay of the oscillations  $\delta$ , which is typical for superconducting transitions.

The experiment conducted under the same conditions the next day shifted the beginning of the feature observed in the dependence  $t(T)$  to  $T = 180$  K. The value of the “spontaneous” rotation was  $\varphi_0 = 5.87^\circ$ , which is tenths of a degree different from the previous experiment.

The measurement results on the third day are shown in Fig. 13. As one can see, this time a sharp transition to the giant pinning region began with  $T = 185$  K (Fig. 13(a)) and  $\varphi_0 = 5.67^\circ$ .

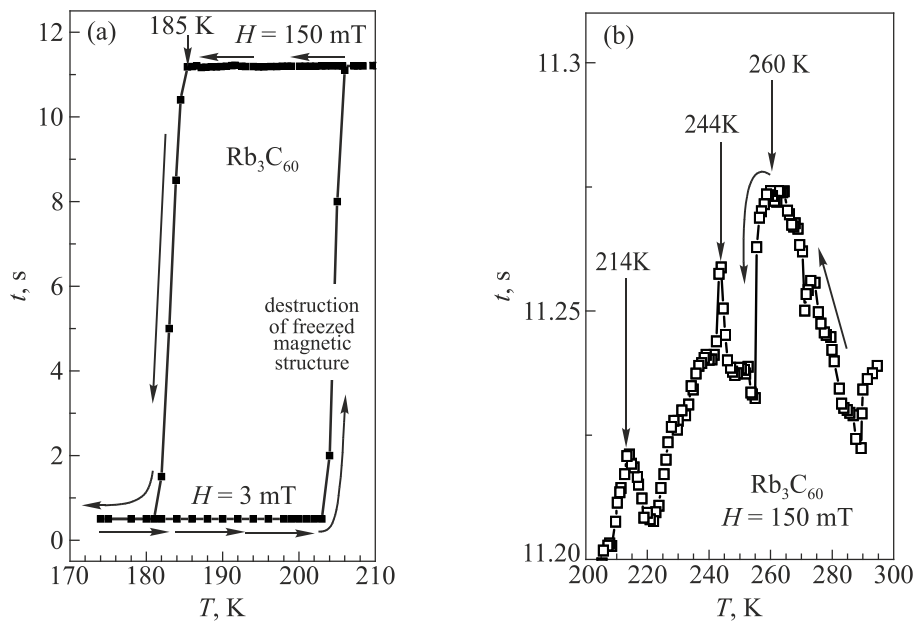
This time, at  $T = 174$  K, the heating furnace was turned on and the magnetic field was turned off at the same time, so that the sample oscillated in the residual field of the electromagnet  $H = 3$  mT. It can be seen that even a decrease in the magnetic field from  $H = 150$  to 3 mT did not change the observed “frozen” picture. The process of heating the  $\text{Rb}_3\text{C}_{60}$  sample in a residual magnetic field lasted

20 min, after which, when  $T = 203$  K was reached, the oscillation period began to grow, and at  $T = 205$  K the normal vibrational mode was restored.

In the same place, Fig. 13(b) shows, on an enlarged scale, a fragment of the temperature region in an experiment with cooling from room temperatures to  $T = 200$  K. As one can see, after the region of the structural  $Fm\bar{3}m-Pa3$  transition at  $T = 260$  K, the oscillation period sharply decreases, thereby showing that the transition is accompanied by magnetic rearrangement. Corresponding to the third experiment dependence of the angle of “spontaneous” rotation  $\varphi_0(T)$  and a schematic representation of the total magnetic moment  $\mathbf{M}$  are shown in Fig. 14.

There are special types of materials for which the cooperative JTE plays a very important role in their structural and magnetic properties. In some compounds, there are ions with orbital degeneracy (the so-called JT ions) and, therefore, in a symmetric structural configuration, ions have not only spin degeneracy, but also additional orbital degeneracy. In these compounds, the crystal lattice is distorted, and their magnetic structure is more complex, having strong anisotropy and magnetostriction. A high concentration of JT ions can lead to JTE cooperation and interchange interactions between them. Such an interaction strongly depends on the spin orientation and can lead to both orbital and spin ordering. We assume that it is precisely this ordering that leads to the effects of giant absorption of oscillations and giant pinning in the region of  $T \sim 200$  K that we observe.

Studies (e.g., a review [44]) of cooperative JTE in various materials show that even at  $T$  higher than the temperatures at which this effect is established, there are local distortions near the JT centers, and they are randomly distributed



*Fig. 13.* Temperature dependences of the oscillation period  $t$  of the  $\text{Rb}_3\text{C}_{60}$  fullerene in the third experiment after annealing, cooled in a magnetic field  $H = 150$  mT: (a) the region of giant pinning with  $T = 185$  K; (b) a fragment of the region of the structural  $Fm\bar{3}m-Pa3$  transition at  $T = 260$  K, on an enlarged scale.

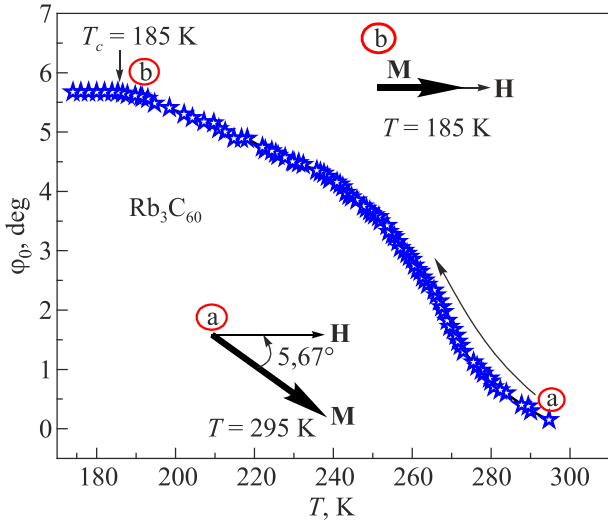


Fig. 14. Temperature dependence of the angle  $\varphi_0$  of the “spontaneous” rotation of the  $\text{Rb}_3\text{C}_{60}$  fullerene, which is cooled to  $T = 174$  K in a magnetic field  $H = 150$  mT, in the third experiment after “annealing”. The insets show schematic images of the direction of the total magnetic moment  $\mathbf{M} = \sum \mathbf{m}_i$ , turning the sample.

over the lattice. On the other hand, distortion of the structure around the JT ion can occur in the  $T^1$  state of fullerite. With decreasing temperature (sample cooling), the number and concentration of  $T^1$  states increase. The intensity ratio  $0:T^1$ , starting at  $T = 400$  K, is 1:2:0, whereas, as shown in [11], this ratio varies significantly in the region of  $T \sim 200$  K and amounts to 35:55:10. At certain temperatures, the density of states  $T^1$  reaches a certain critical value, leading to a cooperative JTE. It can be assumed that the ratio  $T:T^1$

is 55:10, i.e., every sixth  $T$  state is a distorted  $T^1$ , or one JT ion per three  $\text{C}_{60}$  molecules. The estimate shows that for  $T \geq 200$  K, the distance between JT ions is only 2–3 nm, which is quite reasonable for the collective effect to appear in  $\text{A}_3\text{C}_{60}$  fullerenes.

It is interesting to compare the attenuation peaks  $\delta(T)$  from the experiments before the sharp increase ( $A \leq 1^\circ \sim 0.017$  rad), for example, at  $T \geq 160$  K shown in the inset in Fig. 8, with the value up to point (a) at  $T = 225$  K on the same figure and above at the maximum, which is quite difficult to determine, because attenuation increases by several orders of magnitude and falls into an unmeasured region. A comparison of the values of the oscillation period  $t(T)$  before the transition at the points  $T = 211.5$  K (in Figs. 11 and 12), as well as at  $T = 185$  K in Fig. 13(a), with the values after a sharp transition is even more clear. Even a rough estimate of the oscillation frequency  $t = 2\pi/\omega$  (according to the figures given) shows that its change is more than  $\Delta\omega > 10^2$ . From this it can be concluded that during the “transition”, both the pinning force and the number of pinned magnetic dipoles increase, which leads to the state of orbital and spin ordering. As one approaches this state, a giant absorption of oscillations is also observed with a sharp increase in  $\delta(T)$ .

In order to elucidate the physical mechanisms underlying the unusual magnetic properties observed on  $\text{Rb}_3\text{C}_{60}$  fullerene, it is advisable to compare the above results with the detailed (dynamic and static) studies of the magnetic phases of pure  $\text{C}_{60}$  fullerite (99,98% Term. Sublimed) that was studied and described in detail in [44]. Figure 15 shows one of the results of this work, which shows the temperature dependences of the damping decrement  $\delta$  of vibrations of a

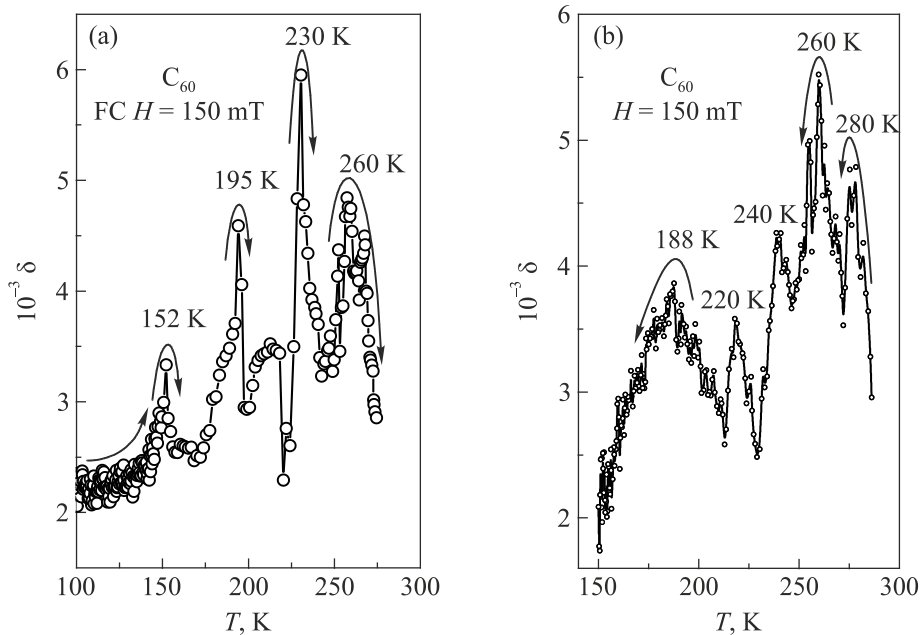


Fig. 15. Temperature dependences of the damping decrement  $\delta$  of vibrations of a  $\text{C}_{60}$  fullerite sample obtained [45] in a transverse magnetic field  $H = 150$  mT under various conditions: (a) after rapid cooling (FC) to  $T = 77$  K and slow heating; (b) in the process of slow cooling from room temperatures to 150 K.

C<sub>60</sub> fullerite sample associated with processes in the rotational subsystem of fullerite molecular rotators.

As we see in Fig. 15(a), a magnetic phase transition is observed in C<sub>60</sub> fullerite at temperatures  $T = 180\text{--}200$  K, with a maximum oscillation damping at  $T = 195$  K, as in the Rb<sub>3</sub>C<sub>60</sub> fullerene (see inset in Fig. 10). Peaks of damping of vibrations in pure fullerite are also clearly visible at temperatures  $T = 152, 230, 260$  K, and they are presumably related to the unusual phenomenon observed in the doped fullerene Rb<sub>3</sub>C<sub>60</sub> which appears, then disappears and “walks” on the temperature scale, the effect of giant absorption of oscillations and (or) giant pinning. Note that after the disappearance, a repeated giant absorption of vibrational energy was observed at  $T \sim 260$  K in the region of the structural transition  $Fm\bar{3}m\text{--}Pa\bar{3}$  and ordering of the axes of rotation of C<sub>60</sub> molecules.

Figure 15(b) shows measurements in a magnetic field  $H = 150$  mT of C<sub>60</sub> fullerite during slow cooling from room temperatures to 150 K, where the attenuation peaks of  $\delta$  oscillations are visible at temperatures of 280, 260, 240, 220 K and a wide peak at  $T = 188$  K. A comparison of the dependences  $\delta(T)$  in Figs. 15(a) and (b) shows that, upon cooling, the attenuation peak more clearly manifests itself in the region of the structural transition  $Fm\bar{3}m\text{--}Pa\bar{3}$  at  $T \sim 260$  K, and the peak at  $T = 220$  K has shifted. As one can see, it is in pure C<sub>60</sub> fullerite that a magnetic phase transition is observed at temperatures  $T = 180\text{--}200$  K, where an unusual gigantic damping of torsion vibrations of the sample and pinning of magnetic dipoles in the Rb<sub>3</sub>C<sub>60</sub> fullerene were discovered, which are very similar in nature to the superconducting transition (see Figs. 11–13 compared with Fig. 2) and after many days of annealing of Rb<sub>3</sub>C<sub>60</sub> fullerene together with the dopant at  $T \geq 40$  °C.

### Conclusions

In this work, in dynamic experiments when studying the magnetic properties of Rb<sub>3</sub>C<sub>60</sub> fullerene in the normal state, both ordinary and not quite ordinary phenomena were observed, such as a giant absorption of oscillations and giant pinning of the magnetic moments of dipoles, depending on the background of the experiment, magnetic field  $\mathbf{H}$ , and sample orientation in this field. Observed at  $T \sim 250\text{--}280$  K features can be compared with the structural transition  $Fm\bar{3}m\text{--}Pa\bar{3}$  and explained by the effect of “freezing” of the free rotation of C<sub>60</sub> molecules. We attribute the phenomena of gigantic absorption of oscillations and giant pinning at  $T \sim 200$  K, which are discussed in the framework of the cooperative Jahn–Teller effect, to not quite ordinary.

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### Незвичайні магнітні явища в динамічних торсіонних дослідженнях фуллерена $\text{Rb}_3\text{C}_{60}$

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В динамічних експериментах з використанням високочутливої торсіонної техніки досліджено магнітні властивості допованого рубідієм монокристалу фуллерита  $\text{Rb}_3\text{C}_{60}$  в широкому інтервалі температур. Визначено та досліджено критичну температуру надпровідного переходу  $T_c$ , критичне магнітне поле  $H_{c1}$ , сили пінінгу як вихорів Абрикосова, так і магнітних моментів диполів при  $T \gg T_c$ , при яких істотні релаксаційні процеси. Показано, що структурний перехід  $Fm3m-Pa3$  при  $T = 260$  К супроводжується реформуванням магнітної структури. Спостерігалися як звичайні при  $T \sim 160-250$  К, так і не зовсім звичайні піки гігантського поглинання енергії осциляцій, які з часом зміщуються до кімнатних

температур. Явище обговорено в рамках кооперативного ефекту Яна–Теллера з урахуванням можливого орбітального та спінового впорядкування. Досліджено релаксаційні процеси при кімнатних температурах ( $T \sim 300$  К). Після багатоденної витримки (відпалу) фуллерену при  $T \geq 40$  °С в експериментах з охолодженням від кімнатної температури при  $T \sim 200$  К виявлено явища, типові для надпровідного переходу з гігантським пінінгом та загасанням осциляцій. Наведено результати досліджень магнітних фазових переходів при  $T = 180-200$  К в чистому фуллериті  $\text{C}_{60}$  (99,98%), які зіставлено з виявленими незвичайними магнітними явищами в фуллерені  $\text{Rb}_3\text{C}_{60}$ .

Ключові слова: фуллерит,  $\text{Rb}_3\text{C}_{60}$ , ефект Яна–Теллера, методика торсіонних коливань, захоплений магнітний потік, молекулярні ротатори.

### Необычные магнитные явления в динамических торсионных исследованиях фуллерена $\text{Rb}_3\text{C}_{60}$

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В динамических экспериментах с использованием высокочувствительной торсионной техники исследованы магнитные свойства допированного рубидием монокристалла фуллерита  $\text{Rb}_3\text{C}_{60}$  в широком интервале температур. Определены и исследованы критическая температура сверхпроводящего перехода  $T_c$ , критическое магнитное поле  $H_{c1}$ , силы пиннинга как вихрей Абрикосова, так и магнитных моментов диполей при  $T \gg T_c$ , при которых существенны и релаксационные процессы. Показано, что структурный переход  $Fm3m-Pa3$  при  $T = 260$  К сопровождается реформацией магнитной структуры. Наблюдались как обычные при  $T \sim 160-250$  К, так и не совсем обычные пики гигантского поглощения энергии осцилляций, со временем смещающиеся к комнатным температурам. Явление обсуждено в рамках кооперативного эффекта Яна–Теллера с учетом возможного орбитального и спинового упорядочения. Исследованы релаксационные процессы при комнатных температурах ( $T \sim 300$  К). После многодневной выдержки (отжига) фуллерена при  $T \geq 40$  °С в экспериментах с охлаждением от комнатной температуры при  $T \sim 200$  К обнаружены явления, типичные для сверхпроводящего перехода с гигантским пиннингом и затуханием осцилляций. Приведены результаты исследований магнитных фазовых переходов при  $T = 180-200$  К в чистом фуллерите  $\text{C}_{60}$  (99,98%), которые сопоставлены с обнаруженными необычными магнитными явлениями в фуллерене  $\text{Rb}_3\text{C}_{60}$ .

Ключевые слова: фуллерит,  $\text{Rb}_3\text{C}_{60}$ , эффект Яна–Теллера, методика торсионных колебаний, захваченный магнитный поток, молекулярные ротаторы.