Triangular lattice antiferromagnet $RbFe(MoO_4)_2$

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A S Borovik-Romanov started working under the guidance of Petr Georgievich Strelkov, and soon grew to be one of the leaders among researchers in the physics of magnetic phenomena. For many years the 'official' head of the 'magnetic diaspora' here was Sergei Vasil'evich Vonsovskii, but after him Andrei Stanislavovich undoubtedly became the leader in researching magnetic phenomena.

A S first gained fame when, together with M P Orlova, he discovered weak ferromagnetism in antiferromagnets. In principle, this phenomenon had already been observed, but at that time it was assumed that it was caused by the presence of uncontrolled impurities producing incomplete compensation of the magnetic moments of two sublattices, and this resulted in a nonzero ferromagnetic moment. What A S and M P Orlova achieved was a demonstration that this phenomenon had no connection to impurities and reflected, so to speak, the nature of things.

Here again, Andrei Stanislavovich was immensely lucky — a close creative contact appeared in the person of theorist Igor Ekhiel'evich Dzyaloshinskii who also worked at our institute (IPP RAS). This collaboration generated numerous very interesting results, both theoretical and experimental. Petr Leonidovich Kapitza always pointed to it as an example of the fruitful cooperation of theory and experiment. The most important result born of it was the prediction and discovery of piezomagnetism. Many books advanced an opinion at the time that piezomagnetism was impossible because strain does not change in response to a reversal of the sign of time, while it reverses the sign of the magnetic moment. This is not true, however, if one is dealing with a state not invariant under time reversal, and any antiferromagnetic state belongs to this class. That was it: Andrei Stanislavovich discovered piezomagnetism in antiferromagnets.

Many outstanding results followed, obtained in collaboration with his students — Lyudmila Andreevna Prozorova, Natalya Mikhailovna Kreinis, and many others, for instance, Aleksandr Ivanovich Smirnov. On the whole, a fairly numerous group of brilliant people working on magnetism grew up in our institute, and they continue working on it.

Now, of course, magnetism is a far cry from what it was in the time of Borovik-Romanov — a great deal of water has flowed under the bridge — but his scientific school continues to occupy a very high level on a global scale, and what it does is precisely modern magnetism. I remember very well Andrei Stanislavovich's reaction to the discovery of the superfluidity of helium-3. That was in 1972. He was very much impressed by this discovery, especially after he realized that, in terms of magnetism, superfluid helium-3 constitutes antiferromagnet. A S then formulated the task of researching superfluid helium-3 precisely as an antiferromagnet. It is worth emphasizing at this point that even as we speak there is still not a single group within the confines of the former Warsaw Pact block which has successfully worked with superfluid helium-3. Note that Borovik-Romanov launched this research in the mid-1980s. He had built up a very creative team of young researchers: Yu M Bunkov, V V Dmitriev, Yu M Mukharskii, D A Sergatskov. Also in this team was Anita de Waard from The Netherlands and other visiting scientists. They succeeded in building a facility which is still unique and capable of achieving temperatures down to a tenth of a millikelvin.

Of high importance was Borovik-Romanov's foresight that the especially interesting features of superfluid helium-3 are not those which made superfluid helium-4 exciting (i.e., superfluidity), but precisely its magnetic properties. As a result, Andrei Stanislavovich's team discovered a novel phenomenon now known as magnetic superfluidity. A theorist also working at our institute, Igor Akindinovich Fomin, played a significant role in the discovery of this phenomenon, as well. The authors called it "homogeneously precessing domain." Nowadays, it is more fashionable to talk about Bose condensates. In these terms, this is a nonstationary Bose condensate, such that the system is in a state of coherent precession in time, while being homogeneous in space.

Andrei Stanislavovich simply loved traveling. I remember how A S and his wife at a Conference in Odessa were carefully studying the map of Odessa's environs and were enthusiastically tracing walks for independent excursions. For A S this passion proved fatal. His wish to attend the International Conference on Magnetism in Australia in July 1997 overpowered the resistance of his doctors, who were adamant that this long flight would be critically dangerous for Andrei Stanislavovich's health. However, A S insisted that he should go; alas, it was his last trip... .

A S's death was totally unexpected for everyone around him: his thoughts were about his beloved science, and he was full of plans and new ideas; alas, it was his colleagues and disciples who had to implement these plans — without Andrei Stanislavovich. The science that A S was doing continues to unfold, both at IPP RAS and in many other laboratories in various countries, as will be discussed today in subsequent research reports during this session.

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Triangular lattice antiferromagnet $RbFe(MoO_4)_2$

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The investigation of the magnetic ordering of spins on a two-dimensional triangular lattice led to the discovery of unordinary phase transitions caused by the frustration of the antiferromagnetic exchange interaction and by the effect of fluctuations. The antiparallel ordering of spins corresponding to the minimum energy of pairwise interactions cannot be realized for a triangular lattice: given the antiparallel orientation of the first and second spins on a triangle, the third spin cannot be directed strictly opposite to both the first and second ones. The minimum of the exchange energy for classical spins ($S \gg 1$) can be realized in a three-sublattice configuration in which on each triangle the spin directions make an angle of 120° with one another [1, 2]. Anderson [3]

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supposed that the spins $S = 1/2$ on a triangular lattice in the ground state are completely disordered and are in the state of a quantum spin liquid. Subsequent theoretical investigations based on a numerical simulation taking account of interaction between only nearest neighbors have demonstrated, however, that a three-sublattice 120° ordering arises in this case as well, but the spin reduction due to quantum zero point fluctuations is very large — the ordered spin component comes to only 40% of the nominal magnitude [4]. In experiments with real magnetic systems having a regular triangular lattice, systems with a 'triangular', i.e., three-sublattice 120° ordering, as well as quantum-disordered systems without a magnetic order, were observed as the temperature decreased to very low values (see, e.g., Ref. [5]). In this report, we consider the problem of an ordered phase of a two-dimensional antiferromagnet with quasiclassical spins on a regular triangular lattice and its phase transformations in a magnetic field.

In a magnetic field, a classical antiferromagnet with a Heisenberg exchange on a triangular lattice possesses degeneracy of a specific type [1], where all spin configurations of the three sublattices with equal total moments have the same energy in the molecular-field approximation, e.g., configurations of the a, b, b′ types in Fig. 1.

The ground state is determined in this case by fluctuations. For an equilibrium system, the free energy with allowance for quantum and thermal fluctuations, which depend on the spectra of spin waves, should be minimum. A theoretical analysis [1, 2] shows that in weak magnetic fields the fluctuations stabilize the planar spin structure (structure 'b' in Fig. 1) in which the spins lie in the plane parallel to the magnetic field. The alternative umbrella-type structure 'a' proves to be energetically unfavorable. In addition, the fluctuations stabilize a collinear spin structure of the 'two up, one down' type ('c' in Fig. 1) in a wide range of magnetic fields in the vicinity of the field equal to one third of the saturation field, $H = H_{sat}/3$. In the molecular-field approximation, such a structure can be realized only in a field $H_{sat}/3$ and does not lead to the development of any peculiarities in the magnetization curve. The allowance for fluctuations predicts the appearance of a magnetization plateau at a level of one third of the total magnetic moment and the existence of an extended (in field) region in the $T-H$ phase diagram corresponding to the collinear phase 'c'. In the region of high fields preceding the saturation field, the fluctuations stabilize the planar structure 'd'. In theoretical work and in numerical simulations (see Refs [6, 7]), the phase diagrams depicted in Fig. 2 were predicted.

Among the experimental investigations of antiferromagnets on triangular lattices, studies of compounds of the ABX_3 type (A = Cs, Rb; B = Ni, Mn, Cu; X = Cl, Br, I) dominated up to recent years. However, the predominant interaction in them is exchange interaction in the direction perpendicular to planes containing triangular plane lattices of magnetic ions, i.e., these systems represent quasi-one-dimensional magnets (see, e.g., Ref. [8]). We shall consider in this report experimental investigations of a quasi-two-dimensional antiferromagnet with a triangular lattice, $RbFe(MoO_4)_2$, and compare the experimental findings with the results of theoretical models. The magnetic structure of $RbFe(MoO_4)_2$ is formed by Fe^{3+} ions ($S = 5/2$, $L = 0$) located in layers with a regular triangular (hexagonal) lattice (Fig. 3). The magnetic layers are separated by nonmagnetic Rb ions and nonmagnetic MoO_4 complexes, which substantiates the validity of the two-dimensional approximation. The moderate magnitude of the main exchange integral permits one to perform investigations in magnetic fields up to the saturation field.

In Fig. 4, the magnetization curves obtained in pulsed magnetic fields of up to 25 T are given [9], and in Fig. 5, in stationary fields up to 10 T [10]. The field was applied in the plane of magnetic layers. The data presented show the existence of a magnetization plateau near $M_{sat}/3$; as is seen from the dM/dH curves, the plateau is expanded with

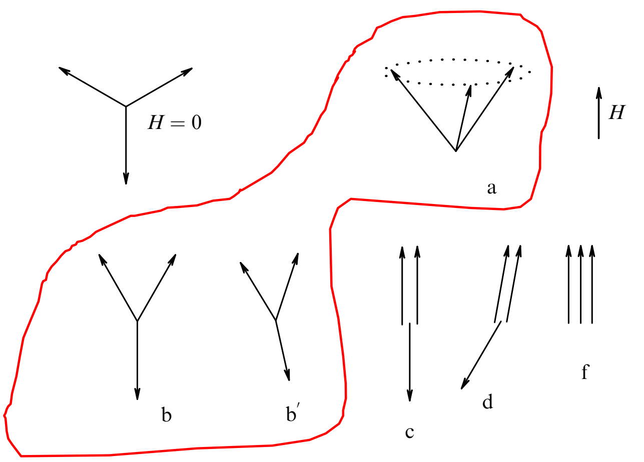

Figure 1. Schematic of magnetic moments of sublattices of a two-dimensional antiferromagnet on a triangular lattice. The outlined configurations correspond to degenerate states with the same total magnetic moment.

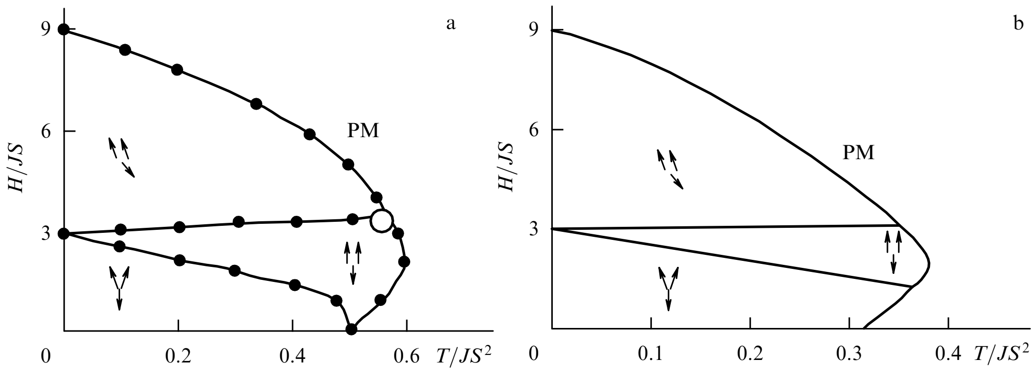

Figure 2. (a) Phase diagram for a two-dimensional XY model [6]. (b) Phase diagram for a two-dimensional Heisenberg model [7]. J is the exchange integral, S is the magnetic ion spin, and PM is the paramagnetic region.

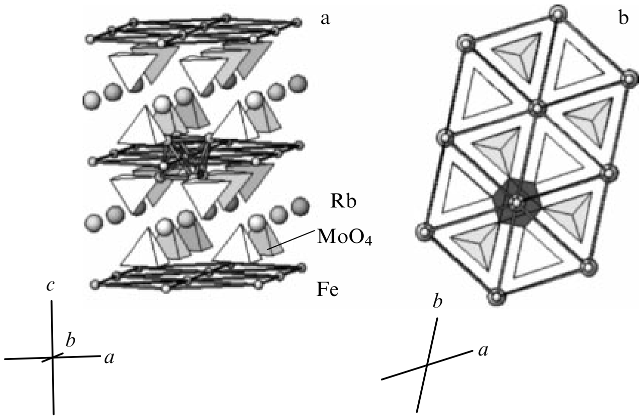

Figure 3. Crystal structure of $RbFe(MoO_4)_2$.

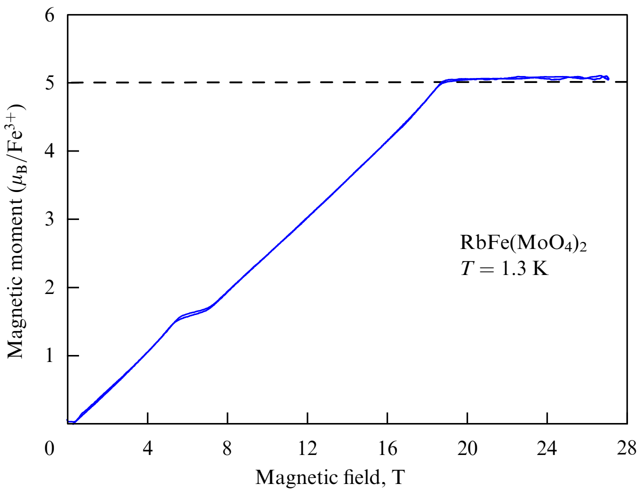

Figure 4. Field dependence of the magnetic moment in the range below 25 T.

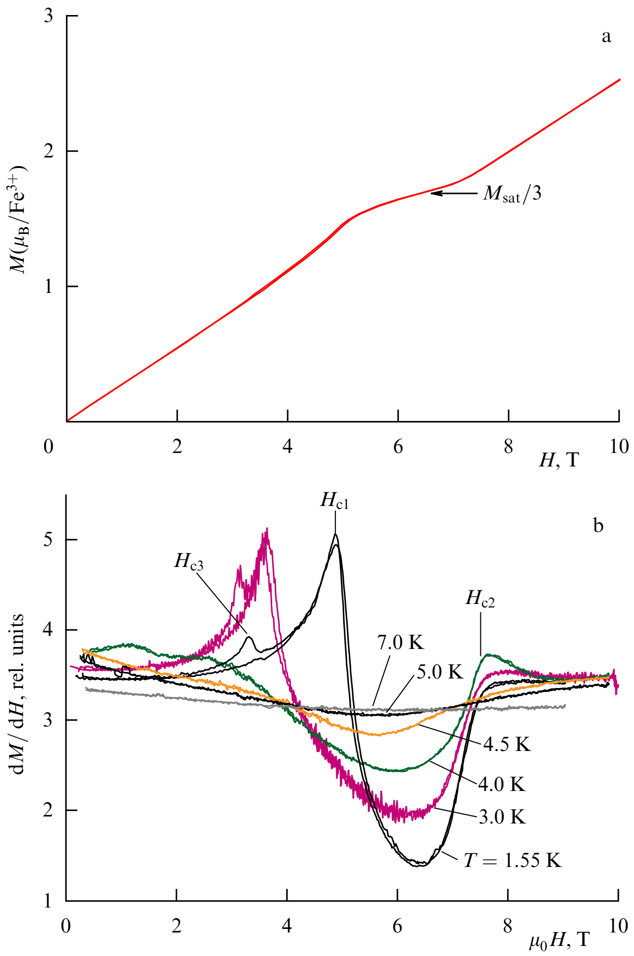

Figure 5. (a) Dependence of the magnetic moment on the field applied in the plane of magnetic layers at $T = 1.55$ K. (b) Field dependence of the derivative dM/dH at different temperatures; H_{c1}, H_{c2}, H_{c3} are the critical fields.

increasing temperature, in accordance with the fact that the thermal fluctuations stabilize the collinear phase. We shall designate the fields that bound the plateau as H_{c1} and H_{c2}. Apart from the values of these two critical fields, the variation of the derivative dM/dH reveals one more peculiarity, whose magnetic field is designated in Fig. 5 as H_{c3}; this field will be discussed below. Along with the magnetization curves, the temperature dependences of the magnetic moment in different fields [11], field and temperature dependences of the specific heat [9, 10], and spectra of electron paramagnetic resonance [9, 11] and nuclear magnetic resonance [10, 12] were also studied. The examples of the temperature dependences of the specific heat are given in Fig. 6. The data on the specific heat give clear evidence of a temperature-induced transition from the paramagnetic phase [a sharp peculiarity in the $C(T)$ curve]. When measuring $C(T)$ dependences, clear features arise in the field H_{c1} corresponding to the lower boundary of the magnetization plateau, whereas near the field of the upper boundary of the plateau the response of the heat capacity is inconspicuous. The heat capacity also exhibits a feature near the field $H_{c4} > H_{c2}$; the corresponding $C(H)$ dependences are given in Ref. [9]. All these features are presented in the phase diagram in Fig. 7.

Neutron-scattering experiments [13] reveal (in a zero field, at a temperature equal to the Néel temperature $T_N = 3.8$ K) the appearance of magnetic Bragg peaks corresponding to the wave vector $\mathbf{q} = (1/3, 1/3, 0.457)$ of the magnetic structure in the units of the reciprocal-lattice vectors of the $RbFe(MoO_4)_2$ crystal. This temperature agrees well with the temperature of the specific-heat peak and of the bend in the magnetic susceptibility curve. The values of the components (q_x, q_y) agree with the formation of a three-sublattice 120° magnetic structure in the magnetic layers of iron ions, and the value of the component q_z indicates the mutual orientation of spins in neighboring layers at an angle of 165°. Thus, the spins of magnetic ions of adjacent planes neighboring along the direction of the z-axis are ordered, deflecting from the strictly antiparallel mutual orientation, and form a spiral with an incommensurate period.

Under the action of the magnetic field applied in the plane of layers of iron ions, a change in the system of Bragg peaks occurs. In a field close to the above-mentioned H_{c3} field, a transition occurs from the incommensurate structure to a commensurate structure with a period $3c$, i.e., with a wave vector $\mathbf{q} = (1/3, 1/3, 1/3)$, and then, in greater fields exceeding the field H_{c4} which was determined when describing heat capacity, a structure with $\mathbf{q} = (1/3, 1/3, 0.41)$, incommensurate along the z-axis, is observed again. It should be noted that the low-field transition from the incommensurate to commensurate structure is likely to occur not exactly in the H_{c3} field but somewhat higher, between the fields H_{c3} and H_{c1}, since the accuracy of determining the field of transition from

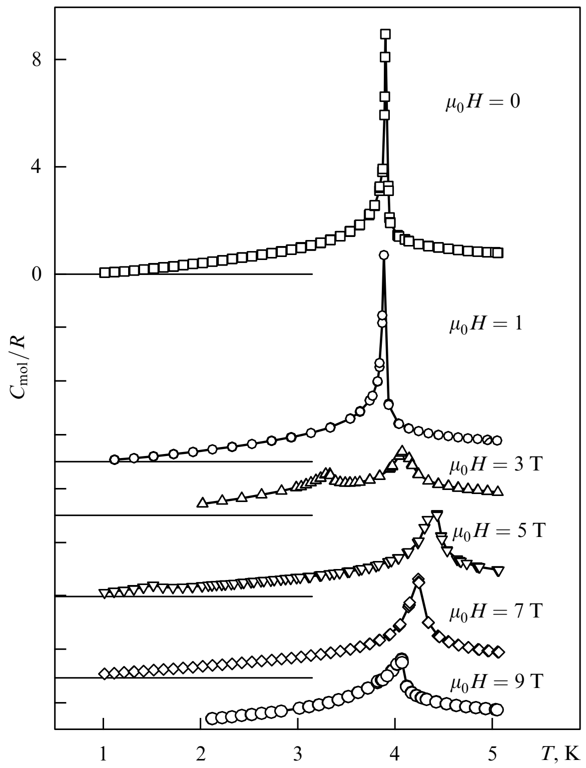

Figure 6. Temperature dependences of the heat capacity in different fields. The field is applied in the plane of magnetic layers, and R is the universal gas constant. For clarity, the curves for the nonzero values of the field are shifted downward.

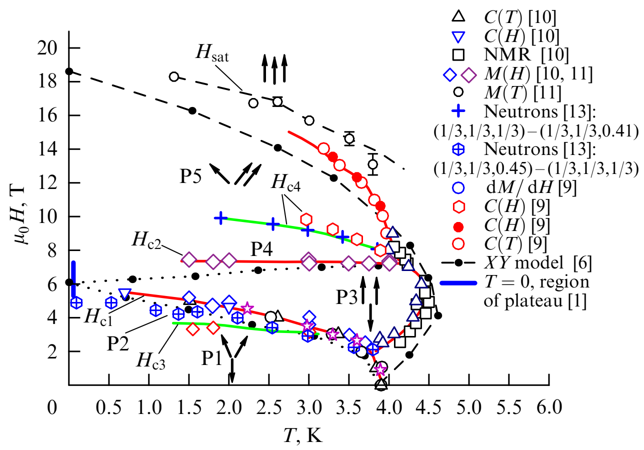

Figure 7. Phase diagram of magnetic states of $RbFe(MoO_4)_2$ for a field directed in the plane of magnetic layers: P1, P5, noncollinear incommensurate phases; P2, noncollinear commensurate three-sublattice phase; P3, collinear phase of the 'two up, one down' type (magnetization plateau), and P4, noncollinear commensurate two-sublattice phase. Arrows illustrate the spin structure of individual magnetic layers in the three-sublattice model.

one wave vector to another appears to only slightly exceed the difference $H_{c1} - H_{c3}$ (the change in the Bragg reflections arises upon a variation in the magnetic field by 1 T).

The transverse components q_x, q_y of the wave vector of the structure do not change with the variation of the magnetic field in the entire range of measurements. The changes in the structure revealed in neutron experiments are marked in the phase diagram (Fig. 7). For the high-field transition (1/3, 1/3, 1/3) $\longrightarrow$ (1/3, 1/3, 0.41), an upper boundary of the region is shown in the figure, where reflections of both types coexist. The alterations in neutron scattering related to the change in the wave vector of the structure do not fix the boundaries of the magnetization plateau (at least its upper boundary), but prove to be quite sensitive to the rearrangement of the mutual orientation of spins in the neighboring layers with respect to each other. With these rearrangements, a small-pronounced peculiarity is observed in the magnetization curve in the H_{c3} field and in the behavior of the specific heat in the H_{c4} field. The temperatures of the bifurcation of the lines of nuclear magnetic resonance (NMR) related to the appearance of an antiferromagnetic order parameter agree well with the temperatures of the specific-heat peak in the entire range of magnetic fields in which the NMR signal was detected. Thus, the different methods of investigation make it possible to determine the phase diagram of $RbFe(MoO_4)_2$ on the (T, H) plane and then to suppose that the phase transformations in fields H_{c1} and H_{c2} are connected with the phases of two-dimensional ordering in 'triangular' layers and that the rearrangement of the structure in the H_{c4} field is connected with the changes in the mutual orientation of spins in neighboring layers, being related to the three-dimensional magnetic structure. The rearrangement of the structure in the H_{c3} field appears to be also caused by changes of the latter type.

In the spectrum of electron paramagnetic resonance studied in Refs [9, 11], a splitting of magnetic resonance modes of $RbFe(MoO_4)_2$ related to the interplane exchange interaction was observed. The ratio of the exchange integrals inside a layer and between the layers that is determined from this splitting is equal to 100, i.e., $RbFe(MoO_4)_2$ is a good realization of a two-dimensional spin system. An analysis of the spectrum of electron spin resonance and the investigation of the anisotropy of the saturation field make it possible to determine the anisotropy type by two independent methods. The $RbFe(MoO_4)_2$ magnetic system has an anisotropy of the easy-plane type; the characteristic anisotropy field is $DS/g\mu_B = (5.7 \pm 0.5)$ kOe according to magnetic resonance data, and $DS/g\mu_B = (8.5 \pm 1.5)$ kOe according to data on the saturation field (here, D is the constant of uniaxial anisotropy, g is the g factor, and the corresponding spin Hamiltonian was given in Ref. [11]).

The discrepancy between the magnitudes of the characteristic anisotropy field, obtained by two different methods, which appears to be caused by the renormalization of the effective anisotropy field due to zero point fluctuations, is discussed in Refs [1, 9]. It can be expected that the behavior of a two-dimensional antiferromagnet on a triangular lattice with a strong easy-plane anisotropy will be described by the classical XY model [6]. In this case, the only parameter of the theoretical model is the exchange field JS. This quantity can be determined in an independent way from the data on the susceptibility or on the saturation field [10].

Figure 7 displays the results of such a theoretical calculation for the transition temperature from a paramagnetic phase to an ordered three-sublattice phase and for the fields H_{c1} and H_{c2} that bound the region of the magnetization plateau $M_{sat}/3$. For this construction, no adjusting parameters have been used; the magnitude of $J = 1.2$ K was taken from the observed value of the saturation field ($H_{sat} = 9JS = 182$ kOe in the notation employed in Refs [6, 7]). The theoretical value of the transition temperature $k_B T_N = 0.5JS^2$ at $S = 5/2$ and $J = 1.2$ K is in good agreement with the observed value of 3.9 K. The boundaries between the collinear and noncollinear phases are also in satisfactory agreement with experiments. An increase in the Néel temperature under the effect of a magnetic field also corresponds to the predictions of the model. Notice that an ordinary antiferromagnet is characterized by the opposite effect of the field on the transition temperature. In the classical model [6] under consideration, the width of the interval of fields in which the fluctuations stabilize the phase with a moment $M_{sat}/3$ tends to zero with decreasing temperature. The allowance for quantum fluctuations should lead to a nonzero interval at the zero temperature. The estimate of the width of this interval [1] caused by quantum fluctuations, which is shown in Fig. 7 by a vertical bar near the ordinate axis, is also in agreement with the extrapolation of the experimental dependences $H_{c1}(T)$ and $H_{c2}(T)$ to the zero temperature. One discrepancy with the predictions of the model is the nonzero value of the field H_{c1} at temperatures immediately adjoining T_N from below. It should be noted that the nonzero value of H_{c1} is predicted on the basis of the Heisenberg model [7]. The high-field phase boundary between the canted antiferromagnetic and paramagnetic phases also demonstrates an unordinary fluctuation behavior. In the region of temperatures exceeding 2 K, the specific-heat peak in the $C(H)$ dependence is observed in a field that is lower than the field of tending to saturation in the magnetization curve, determined from the falloff of the derivative dM/dH. The positions of the singularities in the $C(H, T)$ curves and in the dM/dH field dependences are shown in the phase diagram in Fig. 7 near the high-field boundary of the ordered phase. The discrepancy at $T = 3$ K constitutes about 1 T. The scenario of the two-step transition to the saturated phase for a two-dimensional antiferromagnet on a triangular lattice, which was predicted in Ref. [2], is related to fluctuations: in the lower critical field, the long-range order of spin components perpendicular to the magnetic field disappears. In the interval between the two upper critical fields, the correlation between the transverse spin components falls according to a power law, with the sample remaining unsaturated. In the upper critical field, the correlations begin decreasing according to an exponential law, and the transverse component disappears.

Thus, the $RbFe(MoO_4)_2$ crystal represents a model system corresponding to a classical two-dimensional antiferromagnet on a triangular lattice. The character of the phase diagram and the existence of a magnetization plateau demonstrate good agreement with the results of a theoretical simulation of this system in terms of the classical two-dimensional XY model.

Some aspects of three-dimensional (i.e., interlayer) ordering were beyond the scope of discussion in this report; on a qualitative level they can be considered [10] based on an analysis of the interlayer interaction and related phases, which was performed in the theoretical work [14].

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Spin superfluidity and magnons Bose–Einstein condensation

Yu M Bunkov

The prehistory of the discovery of magnetic superfluidity goes back to the mid-1970s, when two students at Moscow Institute of Physics and Technology (MFTI in *Russ. abbr.*), Boris Dumesh and Yuriy Bunkov, started studying, under the guidance of Academician Andrei Stanislavovich Borovik-Romanov, antiferromagnetic crystals with a dynamic frequency shift. The experiments were mainly performed using $MnCO_3$ and $CsMnF_3$. In these antiferromagnets, the hyperfine field of manganese atoms gives rise to a strong polarization of ^{55}Mn nuclei, such that their frequency of precession becomes about 600 MHz. This frequency is comparable to the frequency of the low-frequency line of antiferromagnetic resonance in a weak external magnetic field. As a result, modes of coupled electron–nucleus oscillations are formed, whose frequency depends on the magnitude of interaction, viz. on the projection of the nuclear magnetic moment onto the magnetization axis of the atoms. The frequency shift of the quasi-NMR of ^{55}Mn nuclei can reach several hundred megahertz at a temperature on the order of 1 K, as is shown in Fig. 1, and decrease upon heating or upon deflection of the magnetization vector of the nuclear subsystem. This results in a strong nonlinearity of the nuclear magnetic resonance (NMR) — the frequency of the precession depends on the angle of deflection of the nuclear magnetization vector. Under these conditions, the effective mechanism of formation of a spin echo is the mechanism of frequency modulation rather than the Hahn echo mechanism. The results of successful investigations of this echo formation mechanism by the researchers of our group were reported in Refs [1–3].

The antiferromagnetic resonance can also be excited parametrically, by the modulation of the external magnetic field at a doubled frequency. It also proved possible to parametrically excite an NMR mode. A new formation mechanism of an echo was discovered, in which the echo was excited by a single resonance pulse and then by a single pulse of parametric pumping [4]. This mechanism of echo

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