Magnetic resonance in the noncollinear antiferromagnet $RbMnBr_3$

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The antiferromagnetic resonance spectrum of the quasi-one-dimensional easy-plane antiferromagnet $RbMnBr_3$ with noncollinear magnetic ordering has been studied. It is shown that in high magnetic fields ($H > 30$ kOe) it is well described by expressions for an antiferromagnet with triangular 120 degree magnetic structure. On applying a magnetic field of 27–30 kOe in the basal plane of the crystal hysteresis phenomena are found in the resonant energy absorption. A symmetry analysis is carried out of the role of spontaneous distortions of the crystal structure in producing the magnetic properties of $RbMnBr_3$.

1. INTRODUCTION

$CsMnBr_3$ and $RbMnBr_3$ are noteworthy among the many quasi-one-dimensional hexagonal antiferromagnets with noncollinear magnetic ordering recently examined in various experimental and theoretical studies. These compounds possess a combination of magnetic properties which is in many respects unique. 1) Peculiarities of the crystal structure make them magnetically quasi-one-dimensional: the Mn^{2+} magnetic ions form chains extending along the hexagonal crystal axis, with the interaction between neighboring spins in the chains much stronger than that of spins belonging to neighboring chains. 2) On lowering the temperature the spin system undergoes a transition into a three-dimensionally ordered antiferromagnetic state in which all the spins lie in the basal plane and the nearest neighbors along the hexagonal axis are antiparallel, while in the basal plane the angle between neighboring spins is 120° or close to this value.

Continuing the analogy between $CsMnBr_3$ and $RbMnBr_3$, we note that in both cases the "easy plane" type of anisotropy exceeds in magnitude the interchain exchange interaction. In $CsMnBr_3$ this leads to a weak field-dependence of the ground state of the spin system.[1] In addition, in the absence of a magnetic field two of the three acoustic modes of antiferromagnetic resonance (AFMR) can be of higher frequency than the AFMR exchange modes.

The magnetic properties of $CsMnBr_3$ are very adequately described by the Hamiltonian

$$\mathscr{H}=2J\sum_{i,j}{}' \mathbf{S}_i\mathbf{S}_j+2J' \sum_{i,j}{}'' \mathbf{S}_i\mathbf{S}_j+D\sum_i (\mathbf{S}_i^z)^2-\mathbf{H}\sum_i \mathbf{S}_i, \tag{1}$$

where $\mathbf{S}_i$ and $\mathbf{S}_j$ are the Mn ion spins. The first term on the right hand side of Eq. (1) describes the strong intrachain exchange interaction (J) of nearest neighbors along the hexagonal axis. The second term describes the weak interchain exchange (J'), with $J \gg J' > 0$. The third and fourth terms in the Hamiltonian describe, respectively, the uniaxial anisotropy ($D > 0$) and the Zeeman energy. In the absence of a magnetic field the ground state of the Hamiltonian (Eq. 1) corresponds to a "triangular" 120-degree antiferromagnetic structure, the evolution of which in a field is similar to that studied theoretically[1] and experimentally.[2]

It could be expected, in principle, that a similar picture would be observed in $RbMnBr_3$ (the lattice constants of these two compounds have similar values), only taking account of numerical corrections for the slightly different values of the parameters of the spin Hamiltonian (Eq. 1). However, the experimental investigation carried out in the present work shows that behavior similar to that of $CsMnBr_3$ is only observed in $RbMnBr_3$ in sufficiently strong magnetic fields and the field dependence of the AFMR spectra are then described fairly well by the Hamiltonian of Eq. (1). In fields less than 27–30 kOe the high-frequency properties of these two crystals are fundamentally different and it is not possible to explain the nature of this difference while remaining within the framework of the model Hamiltonian (Eq. 1). The probable reason for such a difference is the existence of spontaneous distortions of the crystal lattice in $RbMnBr_3$ found in x-ray structural measurements.[3]

2. CRYSTALLINE AND MAGNETIC STRUCTURE OF $RbMnBr_3$

According to x-ray studies[4,5] and the results of powder neutron diffraction,[6] the crystal structure of $RbMnBr_3$, as also of $CsMnBr_3$, is described by the symmetry group D_{6h}^4 ($P6_3/mmc$) and has very close values of the lattice parameters. Glinka *et al.*[6] also studied the magnetic structure of $RbMnBr_3$. They came to the conclusion that $RbMnBr_3$ goes over at a temperature below $T_N = 8.8 \pm 0.1$ K into a three-dimensional ordered magnetic state: all the spins lie in the basal plane of the crystal with neighboring spins along the C_6 axis oriented antiferromagnetically, while in the basal plane the angle between neighboring spins is not 120° (as in other antiferromagnets with a $CsNiCl_3$ crystal structure and an easy-plane anisotropy, among them also $CsMnBr_3$) but $128.5 \pm 1.4°$. The magnetic structure of this crystal thus does not correspond with the crystal structure.

In later x-ray diffraction studies[3] of the crystal structure of $RbMnBr_3$ additional peaks, very weak in intensity, were found which could not arise for the $CsNiCl_3$ type crystal structure. The authors came to the conclusion that $RbMnBr_3$ is isomorphic with $KNiCl_3$: its structure is described by the space group C_{6v}^3 ($P6_3cm$), the elementary cell contains six formula units with parameters $a = 12.924$ Å, $c = 6.547$ Å.

In our view there are weighty reasons to consider that just in that study the crystal structure was correctly deter-

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mined. The point is that as in $KNiCl_3$ so in $RbMnBr_3$ the sizes of the potassium and rubidium ions are too small for filling densely the hexagonal cubo-octahedra of chlorine and bromine ions respectively, which is also the usual reason for distortion of the ideally packed $CsNiCl_3$ type structure.[7] It is known that $KNiCl_3$, crystallizing with hexagonal close-packing, undergoes a structural phase transition on lowering the temperature (560 K) and at room temperature one third of the columns of octahedra are displaced by 0.2–0.7 Å along the C_6 axis as a result of which the KCl_3 layers are corrugated and the cell parameter in the layer plane is increased $\sqrt{3}$ times.

Evidently, such a structural rearrangement also takes place in $RbMnBr_3$. Indirect evidence for this assertion comes from studies of electron–spin resonance spectra in $RbMgBr_3$ crystals doped with Mn^{2+}, Cr^{3+} and Gd^{3+} (Ref. 8) in which two structural phase transitions are observed on lowering the temperature: at $T = 449$ K the transition from $CsNiCl_3$ type ordering to an intermediate form where part of the columns of octahedra are displaced along the C_6 axis in one direction and part in the other, and then at $T \approx 230$ K a transition to the $KNiCl_3$ type structure.

It becomes clear, in view of what has been said above, that the magnetic structure determined from the neutron diffraction picture and based on an incorrect crystal structure may not be realistic. There is no doubt of the closeness of the magnetic structure of $RbMnBr_3$ to the usual triangular 120 degree structure, but here the character of the known small departures from it can only be explained by an analysis of neutron scattering results, starting from true representations of the crystal's space group. On the other hand, it is reasonable to assume that in the absence of structural distortions the magnetic ordering in $RbMnBr_3$ would be just the same as in $CsMnBr_3$. Based on this point of view it is possible, starting only from symmetry considerations, to understand what kind of changes of magnetic properties should lead to spontaneous distortion of the crystal lattice in $RbMnBr_3$.

3. SYMMETRY ANALYSIS

One of the equivalent hexagonal planes is shown in Fig. 1 and the Mn^{2+} magnetic ions in $RbMnBr_3$ are shown. Unlike the undistorted $CsMnBr_3$ structure, the Mn^{2+} ions in $RbMnBr_3$ do not lie strictly in one plane perpendicular to the hexagonal axis. The full circles in Fig. 1 indicate a positive displacement of all the chains of Mn^{2+} ions along the hexagonal z axis, while the open circles correspond to negative displacement. It is evident that such a form of distortion will lead to a lowering of the crystal class from D_{6h} to C_{6v}[1)] and a tripling of the elementary cell, as was noted by Von Fink and Seifert.[3] An elementary symmetry analysis shows that the corresponding distortion of the crystal structure corresponds to an active two-dimensional irreducible representation pertaining to a two-pronged $K13$ star with prongs $\mathbf{k} = \pm\frac{1}{3}(\mathbf{b}_1 + \mathbf{b}_2)$. Owing to the absence of a gradient Lifshitz invariant, such a phase transition is not generally accompanied by the appearance of an incommensurate lattice.

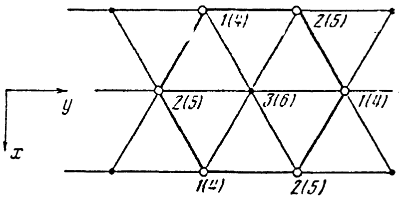

FIG. 1. Distorted crystal structure of $RbMnBr_3$ with space symmetry C^3_{6v}. The numbers in brackets refer to the positions of Mn ions situated at a distance $\pm c/2$ from the plane of the drawing.

The appearance of "triangular" antiferromagnetic ordering in $RbMnBr_3$ does not lead to multiplication of the primitive cell, i.e., in this case the 6-sublattice antiferromagnetic structure corresponds to the coincidence of the magnetic and crystallographic cells. Then, unlike the case of $CsMnBr_3$, six of the magnetic sublattices shown in Fig. 1 decompose into two unconnected sets corresponding to two crystallographically nonequivalent positions of the Mn^{2+} ions:

$$\{S_1, S_2, S_4, S_5\} \text{and} \{S_3, S_6\}. \tag{2}$$

As usual, lowering the crystal symmetry leads to the appearance of additional parameters in the spin Hamiltonian and complicates the structure. In particular, the exchange energy in terms of the sublattice magnetization is determined by the following expression:

$$\begin{aligned}\mathcal{H}_{ex} = {} & J_a(\mathbf{S}_1\mathbf{S}_4+\mathbf{S}_2\mathbf{S}_5)+J_b\mathbf{S}_3\mathbf{S}_6+J_a'(\mathbf{S}_1\mathbf{S}_3+\mathbf{S}_2\mathbf{S}_3+\mathbf{S}_4\mathbf{S}_6+\mathbf{S}_5\mathbf{S}_6)\\ & +J_b'(\mathbf{S}_1\mathbf{S}_2+\mathbf{S}_4\mathbf{S}_5)+J_a''(\mathbf{S}_1\mathbf{S}_6+\mathbf{S}_2\mathbf{S}_6+\mathbf{S}_4\mathbf{S}_3+\mathbf{S}_5\mathbf{S}_3)\\ & +J_b''(\mathbf{S}_1\mathbf{S}_5+\mathbf{S}_2\mathbf{S}_4).\end{aligned} \tag{3}$$

In the absence of spontaneous distortions of the crystal structure we should put in Eq. (3) for the exchange energy

$$J_a=J_b,\ J_a'=J_b',\ J_a''=J_b''. \tag{4}$$

By analogy with Eq. (1) the exchange parameters J and J' in Eqs. (3) and (4) describe, respectively, the intrachain and interchain exchange interactions ($J \gg J' > 0$). As regards the last two terms in the exchange energy, they can be omitted since the parameter J'' describes the clearly negligible exchange interactions of non-closest neighbors.

Two antiferromagnetic states can, in principle, correspond to the minimum exchange energy of Eq. (3) (we consider only the easy-plane phases with sublattice magnetizations with the same modulus):

$$1)\ \text{for}\ J_a' \leqslant 2J_b'\ \cos\varphi_{13}=\cos\varphi_{23}=-J_a'/2J_b', \tag{5}$$

$$2)\ \text{for}\ J_a' \geqslant 2J_b'\ \cos\varphi_{13}=\cos\varphi_{23}=-1. \tag{6}$$

Since we have all grounds for assuming that the exchange constants J_a' and J_b' do not differ very greatly (their difference is proportional to the square of the order parameter of the structural phase transition $D^4_{6h} \rightarrow C^3_{6v}$), it follows that the most probable form of the ground state of $RbMnBr_3$ is given by the solution of Eq. (5) describing the triangular, but now not 120 degree, antiferromagnetic ordering, shown in Fig. 2. In the limiting case of Eq. (4) we have from Eq. (5) $\varphi_{13} = \varphi_{23} = \varphi_{12} = 120°$.

In spite of the fact that the "distortion" of the magnetic structure of Eq. (5) can, according to its parameters, be close to 120 degrees, its symmetry is quite different. Accordingly many of its physical properties, both equilibrium and high-frequency, will be qualitatively different. The most important qualitatively new features of the magnetic properties are listed below, owing their origin to spontaneous distortions of the crystal structure of $RbMnBr_3$.

1. The magnetic structure of Eq. (5), shown in Fig. 2, corresponds to six[2)] types of orientational domains, differeing from one another by rotation of the spins around the hexagonal axis by an angle of 60°. In the absence of structural distortions there are only two types of orientational domains, differing from one another by rotation through 180° around the z axis (or, what is the same, by the time reversal operation $1'$).

2. From the point of view of the paraphase group symmetry (C_{6v}^3), the magnetic structure of Eq. (5) is reducible, two different two-dimensional irreducible representations of the point group C_{6v} take part in its formation. Such a magnetic structure can not, in principle, emerge from the paraphase through a single second order phase transition. The existence of two magnetic phase transitions is then possible with temperatures T_{N1} and T_{N2}. It is significant that the difference between these temperatures is determined by the exchange interactions.

The next three features concern the influence of distortions on the resonance properties of $RbMnBr_3$. We recall first that according to Chubukov[1] and to Vitebskiĭ *et al.*[9] the AFMR spectrum in the undistorted structure consists of three acoustic branches ($a1$, $a2$, $a3$) and three exchange branches ($e1$, $e2$, $e3$). In a zero magnetic field and in the absence of structural distortions the frequencies corresponding to these modes are connected by the relations

$$\omega_{a1} \ll \omega_{a2} = \omega_{a3}, \quad \omega_{e2} = \omega_{e3} \neq \omega_{e1}, \tag{7}$$

where the value of ω_{a1} is only different from zero in line with the presence of a neglibibly small hexagonal anisotropy in the basal plane. The influence of structural distortions leads to the following consequences.

3. All AFMR modes will be non-degenerate even for $H = 0$. The splitting of the frequencies ω_{e2} and ω_{e3} will then be of an exchange nature.

4. The $a1$ acoustic mode acquires a magnetoelastic gap, i.e., a relativistic magnetostriction, quadratic in the spins, takes part in producing the magnitude of ω_{a1}, which would not be so in the absence of structural distortions.

5. The existence of orientational domains (Sec. 1) leads to a complicated splitting of all resonance frequencies in an external magnetic field which is not parallel to the hexagonal crystal axis, since the different domains will be differently oriented relative to the field. The additional splitting will only disappear on reaching an external field H stronger than the characteristic field for switching the domain structure.

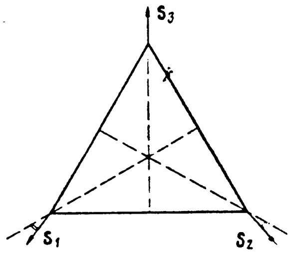

FIG. 2. Triangular antiferromagnetic structure in a distorted crystal lattice. Spins in positions *4*, *5* and *6* are antiparallel to spins in positions *1*, *2* and *3*, respectively. The orientation of the magnetic structure as a whole in the xy plane is shown as arbitrary.

Summarizing the five points made above, we note that spontaneous disortions of the crystal structure, the existence of which is assumed in $RbMnBr_3$, should actually lead to fundamental changes in the magnetic properties of this compound—this is unavoidable as a consequence of symmetry. However, we do not have available any information on the quantitative aspect of the problem. We only note that the appearance of structural distortions will in fact lead to a doubling of the number of independent spin-Hamiltonian parameters contained in expressions for the AFMR frequencies.

6. So far we have tacitly assumed that the crystal structure of $RbMnBr_3$ is sufficiently perfect. However, results of x-ray structural analysis[3] rather point to the opposite, that is to say that the structural phase transition $D_{6h}^4 \rightarrow C_{6v}^3$ is accompanied by the appearance of crystalline domains of very small dimensions. It is possible that the sizes of these domains amounts to several lattice constants. In that situation one should expect strong inhomogeneous broadening of an AFMR line and also hysteresis phenomena in a magnetic field.

4. EXPERIMENTAL RESULTS

Measurements of the AFMR spectrum of $RbMnBr_3$ were carried out on a direct amplification spectrometer by the usual method: the resonant power absorption was recorded by the change in amplitude of the vhf signal P_{vhf}, of constant frequency ω, passing through a cell containing the specimen, when changing the d.c. magnetic field smoothly. A short-circuited four or eight millimeter waveguide with cylindrical or rectangular resonator was used as an absorption cell. A magnetic field up to 54 kOe was provided by a superconducting solenoid. $RbMnBr_3$ crystals are highly hygroscopic and rapidly transform in air to a white substance which is probably $RbMnBr_3 \cdot 2H_2O$. Therefore, for each experiment the specimen was cut from the middle of a single-crystal block and rapidly coated with polystyrene paste. On drying the paste, a thin protective layer was formed, keeping the specimen from hydration. However, we were unable to prevent hydration completely and as a result of this, on nearly all the $P_{\text{vhf}}(H)$ curves a weak parasitic resonance absorption is visible. The position of this resonance is independent of temperature and magnetic field orientation and obeys the relation $\omega = \gamma H$ with $\gamma = 2.8$ GHz/kOe and evidently corresponds to paramagnetic resonance in $RbMnRb_3 \cdot 2H_2O$.

The results of an experimental investigation of the AFMR spectrum in $RbMnBr_3$ at a temperature $T = 1.3$ K ($T_N = 8.8$ K) (Ref. 6) are shown in Fig. 3. On directing the magnetic field perpendicular to the hexagonal crystal axis (the direction in the basal plane was not fixed) the two most low-lying of the predicted six resonance branches are observed (Fig. 3,a): the relativistic ω_{a1} increasing with field and the exchange ω_{e2} falling, vanishing in the phase transition field H_c, corresponding to the spin flop of the two pairs of sublattices. We note that the intensity of the exchange branch of oscillations falls sharply on directing the field ac-

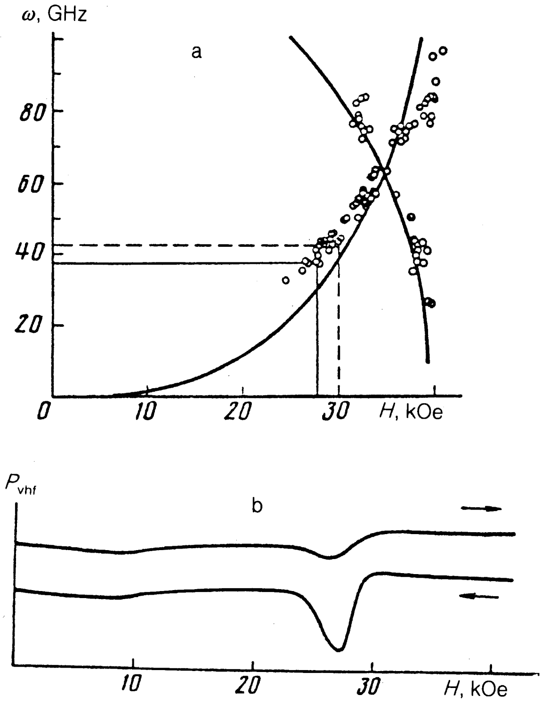

FIG. 3. a—AFMR spectrum in $RbMnBr_3$ at $T = 1.3$ K, field perpendicular to the C_6 axis. The regions of resonance frequencies are shown where "hysteresis" phenomena are observed. b—Example of the trace of the magnetic field dependence of the vhf signal power passing through the resonator containing the specimen for a frequency $\omega = 36.7$ GHz.

curately perpendicular to the hexagonal crystal axis and it was necessary to rotate the dc field from the basal plane by an angle of 2–3° for its observation.

For comparison, the solid lines of Fig. 3a show results of a calculation of the AFMR spectrum in a triangular antiferromagnet with an undistorted crystal structure,[1] for which the parameters $J = 91$ kOe , $J' = 0.057$ kOe, $D = 0.5$ kOe are taken from magnetization measurements. The measurements were initially carried out in the frequency range 42–98 GHz. As can be seen in the figure, the resonance branch decreasing with field agrees well with the ω_{e2} mode, at the same time as the increasing vibrational branch departs slightly from the theoretically predicted dependence for the mode ω_{a1}, especially in weak fields. In order to elucidate the behavior of this branch in weak fields we lowered the frequency and observed the following nontrivial phenomena.

Starting down from 42.5 GHz, i.e. in fields of 30 kOe and less, on recording the microwave signal passing through the absorption cell containing the specimen as a function of magnetic field, the intensity of the absorption corresponding to the mode ω_{a1} in a decreasing dc field was appreciably greater than the absorption intensity in an increasing field (Fig. 3,b). Such an intensity "hysteresis" is observed only for the mode which increases with the field (the intensities of the falling AFMR branch and of the parasitic paramagnetic resonance produced by hydration of the specimen are identical when recorded in either direction) and is not accompanied by hysteresis of the magnetic-field position. On further lowering of the frequency, the resonance line-shape is distorted, becoming asymmetrical and hysteresis of the resonance magnetic field is observed starting from a frequency of 37.3 GHz, i.e., in fields $H < 28$ kOe. On further reducing the frequency the absorption amplitude decreases noticeably without appreciable line broadening, and starting at a frequency of 29 GHz the absorption disappears entirely. We did not find any resonant energy absorption in fields less than 25 kOe, except for the parasitic paramagnetic resonance in the whole available range from 22 to 98 GHz. In addition, we carried out further measurements at 8.9 GHz and also found no traces of AFMR apart from an absorption signal asymmetrical in fields of 27–30 kOe, with hysteresis relative to position in the magnetic field. The absence of resonance in the certainly ordered magnetic structure in such a wide frequency range is, in our view, an extremely interesting and unusual phenomenon.

5. CONCLUSIONS

The experimental results show unequivocally that the high-frequency magnetic properties of $RbMnBr_3$ in high magnetic fields ($H > 30$ kOe) are similar to the properties of the related noncollinear antiferromagnet $CsMnBr_3$ with a 120 degree triangular ordering: both have resonance branches well described by formulae for an AFMR spectrum in a triangular ferromagnet with undistorted crystal structure. Their properties are fundamentally different in lower fields: apart from the departure of the relativistic resonance branch from the theoretical dependence, which can be explained by the appearance of a rather appreciable magnetoelastic gap, unusual hysteresis phenomena are observed and also a sharp reduction down to total disappearance of the resonance amplitude on reducing the frequency.

Our symmetry analysis, taking account of the spontaneous distortions of the crystal structure recorded in $RbMnBr_3$, also leads to the conclusion that the magnetic properties of $RbMnBr_3$ and $CsMnBr_3$, are appreciably different. The results of this analysis are certainly qualitative and do not claim to be a complete description of the behavior of the given magnetic structure in an external field. The main obstacle to a more thorough theoretical analysis is the absence of information about how strongly the parameters of the interchain exchange interaction change upon distortion of the crystal lattice.

One can try to associate the small hysteresis phenomena in the field interval 27–30 kOe with the existence of oriented magnetic domains. To explain the observed sharp reduction in resonance amplitude on lowering the frequency rather than a splitting into several (according to the number of domains) absorption lines, one must assume the existence of a large number of randomly oriented domains produced by the imperfection of the crystal structure. In this case, on reducing the magnetic field the transition from a single-domain state to a fine-grain structure should be accompanied by a sharp reduction in the amplitude of the AFMR line with simultaneous broadening, but such a broadening is not observed experimentally. Apart from this there is no explanation, within the framework of this model, for the sharp reduction in the intensity of several elastic neutron-scattering magnetic peaks when a field of 20–30 kOe is applied in the basal plane, as observed recently by Kawano *et al.*[10]

In our view it is impossible in this connection to confirm definitely the hypothesis of the existence, in weak fields, of a hexagonal magnetic structure in $RbMnBr_3$ incommensurate with the crystal structure.[6] The whole group of phenomena in fields of 27–30 kOe can then be treated as a phase transition with a change of the period of the magnetic structure. The absence of a resonance absorption line in weak fields and the neutron data[10] serve as evidence for this assumption.

Further investigations are necessary for a final elucidation of the physical picture. In particular, an attempt to recalculate the neutron diffraction data with allowance for spontaneous distortions of crystal structure could be highly productive.

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[1)] According to the symmetry classification this is not an intrinsic ferroelectric phase transition. The arising electric polarization is directed along the hexagonal axis.

[2)] For a less symmetrical orientation of a magnetic structure in the basal plane (compare with Fig. 2) the number of orientational domains is doubled.

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