The relaxation mechanism proposed in this article for the formation of the giant echo cannot claim to explain its properties fully. At present, however, it is the only possible feature of the dynamics of spins with DFS which can lead to so appreciable a deviation of the properties of the giant echo from the developed theory of FM echo in systems with DFS. A possible alternative of the relaxation mechanism is participation of other resonant systems in the formation of the giant echo.

It must be noted that systems with coupled electron-nuclear precession are analogous to a number of other systems in which an external action leads to the onset of coherent states. Examples are the coherent state of paramagnetic impurities under the action of an RF field[12] or the superradiant transitions in laser systems.[13] The echo-signal formation mechanism proposed in this article and connected with induced violation of coherence can also be effective in those systems.

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[1)]A recent paper by Tsifrinovich and Krasnov[4] reports an investigation of the influence of the DFS on the microinhomogeneous broadening for a line with a Lorentz shape. Their results, however, agree poorly with the experimental data, possibly as a result of the choice of the form of the broadening.

[2)]The authors thank S. V. Petrov for supplying the crystals.

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Exact solution of the one-ion problem for a magnet with one-ion anisotropy in a field of arbitrary direction

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An exact quantum-mechanical solution is obtained for the one-ion problem for a magnet with one-ion isotropy, located in a magnetic field of arbitrary direction, for a spin $S = 1$. The solution is based on the formalism of the theory of unitary symmetry, in particular on the properties of $SU(3)$ Lie algebra, to which belong the operators that characterize the state of an individual ion. After a number of exact transformation, the one-ion Hamiltonian is reduced to a form for which the determination of the eigenvalues and eigenvectors, of the partition function, of the magnetic susceptibility, of the thermodynamic functions, and others is a trivial problem.

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INTRODUCTION

The simplest model of a system consisting of magnetic ions in lattice sites, in the presence of one-ion anisotropy (OA) and of a magnetic field of arbitrary direction, is described by the Hamiltonian

$$\mathcal{H}=\mathcal{H}_1+\mathcal{H}_{23}, \quad \mathcal{H}_1=\mathcal{H}^a+\mathcal{H}^h;$$
$$\mathcal{H}^a=d\sum_i (S_i^z)^2+e\sum_i [(S_i^x)^2-(S_i^y)^2],$$
$$\mathcal{H}^h=-h_\parallel\sum_i S_i^z-h_\perp\sum_i S_i^x, \quad \mathcal{H}_{23}=\sum_{\substack{ij\\ \alpha\beta=x,y,z}} J_{ij}^{\alpha\beta}S_i^\alpha S_j^\beta. \qquad (1)$$

$h_\parallel$ and $h_\perp$ in (1) are the magnetic-field components parallel and perpendicular to the z axis; for simplicity, the perpendicular component is assumed directed along the x axis. The one-ion anisotropy described by $\mathcal{H}^a$ takes into account the presence of a crystal field that distorts the states of the isolated ion, and the spin-orbit and spin-spin couplings of the electrons of the magnetic ion in the site i. In the case $S=1$, the Hamiltonian $\mathcal{H}^a$ written above has the most general form ($\mathcal{H}^a$ is constant at $S=\frac{1}{2}$). At $S>1$ operators of higher degrees

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in the spin operators can appear.

If the density of the magnetic ions is low and therefore $J \ll d$, an important characteristic of the system are the eigenvalues and eigenvectors of the one-ion Hamiltonian $\mathcal{H}_1$. For example, the EPR and EAR spectra are determined by the energy levels of $\mathcal{H}_1$. When Frenkel magnetic excitons in such systems are investigated, the eigenvectors of the one-ion problem are the basis for the construction of the exciton Hamiltonian. The solution of the one-ion problem is at any rate a necessary stage in the investigation of any characteristic of such a system.

The energy levels and their pertinent eigenvectors of the Hamiltonian $\mathcal{H}_1$ can be easily obtained when the magnetic field is parallel to the anisotropy axis. In this case the secular equation of degree $2S+1$ breaks up onto equations that are quadratic for half-integer S and quadratic and linear for interger S. On the other hand, in the case of an arbitrary direction, the one-ion problem has not been solved and, as noted repeatedly in the literature, difficulties arise when it comes to describing the EPR spectra,[1] to construct a theory of magnetic excitons,[2] etc. In these cases it was necessary to confine oneself to the use of perturbation theory in terms of the field or of the angle between h and the z axis, or to the use of a two-level scheme ($S^{\text{eff}} = \frac{1}{2}$).

Similar difficulties arise when higher-concentrated magnetic systems ($J \gtrsim d$) with OA are used, which admit of ferromagnetic (or antiferromagnetic ordering at low temperatures. In the simplest (molecular-field) approximation the problem reduces to the one-ion problem, and the same difficulties are encountered in the presence of a field of arbitrary direction. These investigations were confined therefore to "trivial" cases $\mathbf{h} \| z$ at $d<0$ ("easy axis" of the OA type,[3] and $\mathbf{h} \perp z$ at $d>0$ ("easy plane" of the OA type;[4] perturbation theory was also used.[5] The problem of taking exact account of the OA in the presence of a transverse field and at $d<0$ (in the molecular field approximation relative to the exchange interaction) was posed in Ref. 6. The approach proposed, however, was limited to the case of zero temperature and $\varepsilon = 0$.

In this paper we solve the one-ion problem in the presence of one-ion anisotropy of $\mathcal{H}^a$ of type (1) (the signs of d and ε are immaterial) and of a magnetic field of arbitrary direction, using unitary-symmetry formalism, particularly the properties of the SU(3) Lie algebra (see e.g., Ref. 7) for $S=1$, and indicate a method for solving the problem at arbitrary S.

SYMMETRY OF $SU(3)$ ALGEBRA AND HAMILTONIAN OF THE PROBLEM

In the case $S=1$ the ion can have three states, and all possible transitions between them are described by eight independent operators belonging to the ASU(3) algebra.[8] They are linearly connected with operators of two types—linear and quadratic in the spin operators. It is convenient to choose the generators of this algebra in our case to be the zero-trace operators O_l^m (l is the rank of the tensor and $m = 0, \pm 1, \ldots, \pm l$):

$$O_1^m = S^m,\ S^{\pm 1} \equiv S^{\pm} = \mp 2^{-1/2}(S^x \pm iS^y),\ S^0 \equiv S^z,$$
$$O_2^0 = (S^z)^2 - \frac{1}{3}S(S+1),\quad O_2^{\pm 1} = -(S^zS^{\pm} + S^{\pm}S^z),\quad O_2^{\pm 2} = (S^{\pm})^2. \tag{2}$$

The tensor operators O_l^m realize the irreducible representations of the group of 3-dimensional rotations. At $l=1$ they coincide with the spin operators. They are also convenient because of the simple commutation relations (without cumbersome coefficients) and their clear meaning, namely: the operators O_l^m are connected with transitions that change the projection of the spin on the z axis by m.

We present the representation of the operators O_l^m in a cyclic basis, which we shall need later on[1)]

$$S^z = \begin{pmatrix} 1 & 0 & 0 \\ 0 & 0 & 0 \\ 0 & 0 & -1 \end{pmatrix},\quad S^+ = \begin{pmatrix} 0 & -1 & 0 \\ 0 & 0 & -1 \\ 0 & 0 & 0 \end{pmatrix},\quad S^- = \begin{pmatrix} 0 & 0 & 0 \\ 1 & 0 & 0 \\ 0 & 1 & 0 \end{pmatrix},$$
$$O^0 = \frac{1}{3}\begin{pmatrix} 1 & 0 & 0 \\ 0 & -2 & 0 \\ 0 & 0 & 1 \end{pmatrix},\quad O^1 = \begin{pmatrix} 0 & 1 & 0 \\ 0 & 0 & -1 \\ 0 & 0 & 0 \end{pmatrix},\quad O^{-1} = \begin{pmatrix} 0 & 0 & 0 \\ -1 & 0 & 0 \\ 0 & 1 & 0 \end{pmatrix}, \tag{3}$$
$$O^2 = \begin{pmatrix} 0 & 0 & 1 \\ 0 & 0 & 0 \\ 0 & 0 & 0 \end{pmatrix},\quad O^{-2} = \begin{pmatrix} 0 & 0 & 0 \\ 0 & 0 & 0 \\ 1 & 0 & 0 \end{pmatrix}.$$

With this choice of the generators, the initial one-ion Hamiltonian takes the form (accurate to a constant that is immaterial in this case)

$$\mathcal{H}_1 = d\sum_i O_i^0 + \varepsilon \sum_i (O_i^2 + O_i^{-2}) + 2^{-1/2}h_{\perp}\sum_i (S_i^+ - S_i^-) - h_{\parallel}\sum_i S_i^z. \tag{4}$$

The rank of the $SU(3)$ algebra is equal to two. This means that its generators (3) include two linearly independent commuting operators. In our case these are the operators S^z and O^0. Any other operator is unitarily equivalent to some linear combination of them.[7] At our choice of the generators, all six independent Hermitian operators, viz. $S^+ - S^-$, $O^1 - O^{-1}$, $O^{-2} - O^{-2}$, $-i(S^+ + S^-)$, $-i(O^1 + O^{-1})$, and $-i(O^2 - O^{-2})$ are unitarily equivalent to the operator S_g. It is clear therefore that any operator A of the considered space can be represented in a certain basis in the form

$$A = \alpha\sum_i S_i^z + \beta\sum_i O_i^0. \tag{5}$$

Our problem is to find such a basis for the Hamiltonian $\mathcal{H}_1$.

This problem is solved in principle by carrying out all the unitary transformations generated by the foregoing six operators:

$$\exp[\varphi_1(S^+ + S^-)],\ \exp[\varphi_2(O^1 + O^{-1})],\ \exp[\varphi_3(O^2 - O^{-2})],$$
$$\exp[i\varphi_4(S^+ - S^-)],\quad \exp[i\varphi_5(O^1 - O^{-1})],\quad \exp[i\varphi_6(O^2 + O^{-2})]$$

and by determining the transformation parameters φ_i from the condition that the corresponding terms of the Hamiltonian vanish. It is necessary for this purpose to establish the explicit form of the matrices of the indicated unitary transformations and the law of transformation of the operators O_l^m,

If, as in our case, the initial Hamiltonian $\mathcal{H}_1$ does not have a y-component of the field, it suffices to carry out three of the six unitary transformations, namely[2)]:

$$V_1=\exp[2^{-1/2}\varphi(S^+-S^-)],\quad V_2=\exp[2^{-1/2}K(O^1+O^{-1})],\quad V_3=\exp[L(O^2-O^{-2})]. \tag{6}$$

UNITARY TRANSFORMATION

The first transformation is well known. It is the rotation through an angle φ about the y axis in 3-dimensional space. The operators O_l^m are then transformed, as is well known, in accord with $\hat{D}^l$-irreducible representations of dimensionality $2l+1$:

$$O_l^m=\sum_{m'}D_l^{mm'}(0,\varphi,0)O_l^{m'}, \tag{7}$$

where $D_l^{mm'}$ is the Wigner D-function.[9] The law of transformation of the operators O_l^m for the remaining two unitary transformations can be obtained in two ways. The first is the explicit construction of the matrices V_i and using (3) on the basis of the Cayley-Hamilton theorem or the Sylvester theorem[10] and next finding directly the $V_iO_l^mV_i$ transformation laws. According to the Sylvester theorem

$$f(\mathrm{A})=\sum_{k=1}^{n}f(\lambda_k)\prod_{i\neq k}(\mathrm{A}-\lambda_i\mathrm{I})\Big/\prod_{i\neq k}\lambda_k-\lambda_i,$$

where n is the order of the matrix A, λ_i are the eigenvalues of A, and I is a unit matrix. By this method we have obtained V_3:

$$V_3=\begin{pmatrix}\cos L & 0 & \sin L\\ 0 & 1 & 0\\ -\sin L & 0 & \cos L\end{pmatrix}. \tag{8}$$

All the operators O_l^m are then transformed as follows:

$$\begin{aligned}&\tilde{S}^z=S^z\cos 2L-(O^2+O^{-2})\sin 2L,\quad \tilde{O}^0=O^0,\\ &\tilde{O}^2=\frac{1}{2}S^z\sin 2L+O^2\cos^2 L-O^{-2}\sin^2 L,\\ &S^+=S^+\cos L+O^{-1}\sin L,\quad \tilde{O}^1=-S^-\sin L+O^1\cos L,\\ &\tilde{O}_l^{-m}=(-1)^m(\tilde{O}_l^m)^+.\end{aligned} \tag{9}$$

We can proceed similarly with respect to V_2. There is, however, a simpler method. It is seen from (9) that at $L=\pi/2$ we have the following unitary transformation of the vectors:

$$\begin{vmatrix}S^+\\ S^z\\ S^-\end{vmatrix}\to\begin{vmatrix}O^{-1}\\ -S^z\\ O^1\end{vmatrix},\qquad \begin{vmatrix}\tilde{O}^2\\ \tilde{O}^1\\ \tilde{O}^0\\ \tilde{O}^{-1}\\ \tilde{O}^{-2}\end{vmatrix}\to\begin{vmatrix}-O^{-2}\\ -S^-\\ O^0\\ -S^+\\ -O^2\end{vmatrix}, \tag{10}$$

so that we can form a subalgebra isomorphic to $ASU(2)$ (or, equivalently, to AO(3)] by the triad of operators O^1, $-S^z$, and O^{-1} in place of S^+, S^z, and S^- as usual. The unitary transformation generated on this algebra and analogous to $\exp[2^{-1/2}\varphi(S^++S^-)]$ is then $\exp(2^{-1/2}\varphi(O^1+O^{-1})]$. The operator transformation law is the same in this case as in the V_1 transformation. Its matrix form is

$$\tilde{\mathrm{A}}=\hat{D}_1\mathrm{A},\quad \tilde{\mathrm{B}}=\hat{D}_2\mathrm{B},\qquad \mathrm{A}=\begin{vmatrix}O^{-1}\\ -S^z\\ O^1\end{vmatrix},\quad \mathrm{B}=\begin{vmatrix}-O^{-2}\\ -S^-\\ O^0\\ -S^+\\ -O^2\end{vmatrix}, \tag{11}$$

D_1 and D_2 are Wigner D matrices. (Their explicit form is too cumbersome to be presented here.) The explicit form of V_2 is

$$V_2=\begin{pmatrix}{}^1/_2(1+\cos K) & -2^{-1/2}\sin K & {}^1/_2(\cos K-1)\\ 2^{-1/2}\sin K & \cos K & 2^{-1/2}\sin K\\ {}^1/_2(\cos K-1) & -2^{-1/2}\sin K & {}^1/_2(1+\cos K)\end{pmatrix}. \tag{12}$$

Thus, after making all three unitary transformations we obtain on the basis of Eqs. (7), (9), and (11)

$$\begin{aligned}\tilde{\mathscr{H}}_1(\varphi,K,L)&=V_3V_2V_1\mathscr{H}_1V_1^{-1}V_2^{-1}V_3^{-1}\\ &=-h(\varphi,K,L)\sum_i S_i^z+d(\varphi,K)\sum_i O_i^0;\end{aligned} \tag{13}$$

$$\begin{aligned}h(\varphi,K,L)&=H(\varphi,K)\cos 2L-D_2(\varphi,K)\sin 2L,\\ H(\varphi,K)&=(h_\perp\sin\varphi+h_\parallel\cos\varphi)\cos K-(d-\varepsilon)\sin\varphi\cos\varphi\sin K,\\ D_2(\varphi,K)&=(h_\perp\cos\varphi-h_\parallel\sin\varphi)\sin K\cos K\\ &\quad+{}^1/_2d(1-2\cos^2\varphi+\cos^2K\cos^2\varphi)\\ &\quad+{}^1/_2\varepsilon(2\cos^2\varphi+2\cos^2K-1-\cos^2\varphi\cos^2K),\\ d(\varphi,K)&=3(h_\perp\cos\varphi-h_\parallel\sin\varphi)\sin K\cos K\\ &\quad+{}^1/_2d(3\cos^2\varphi\cos^2K-1)+{}^3/_2\varepsilon(2\cos^2K-1-\cos^2K\cos^2\varphi).\end{aligned} \tag{14}$$

The angles φ, K, and L are determined from the condition that the terms proportional to S^+-S^-, O^1-O^{-1}, and O^2+O^{-2} vanish in the Hamiltonian, i.e., from the system of equations

$$\begin{aligned}&l(\varphi,K)=0,\quad m(\varphi,K)=0,\\ &H(\varphi,K)\sin 2L+D_2(\varphi,K)\cos 2L=0;\\ &l(\varphi,K)=(\varepsilon-d)\sin\varphi\cos\varphi\cos K-(h_\parallel\cos\varphi+h_\perp\sin\varphi)\sin K,\\ &m(\varphi,K)=-[d\cos^2\varphi+\varepsilon(1+\sin^2\varphi)]\sin K\cos K\\ &\quad+(h_\perp\cos\varphi-h_\parallel\sin\varphi)\cos 2K.\end{aligned} \tag{15}$$

We note that although the initial Hamiltonian did not have terms containing O^1-O^{-1}, it is necessary to carry out all three transformations, for the performance of even the first of them (V_1) leads to the appearance of all the operators from $ASU(3)$.

The determination of the eigenvalues and of the eigenvectors of $\mathscr{H}_1$ is now trivial. The eigenvalues are [see Eqs. (3)]

$$\begin{aligned}&E_1=-h(\varphi,K,L)+{}^1/_3d(\varphi,K),\quad E_2=-{}^2/_3d(\varphi,K),\\ &E_3=h(\varphi,K,L)+{}^1/_3d(\varphi,K).\end{aligned} \tag{16}$$

The eigenvectors in the new basis are eigenvectors of the operator S^z, i.e.,

$$\mathrm{l}_1=\begin{vmatrix}1\\ 0\\ 0\end{vmatrix},\quad \mathrm{l}_2=\begin{vmatrix}0\\ 1\\ 0\end{vmatrix},\quad \mathrm{l}_3=\begin{vmatrix}0\\ 0\\ 1\end{vmatrix}.$$

The eigenvectors $\tilde{\mathrm{l}}_i$ in the initial basis are specified by a matrix that diagonalizes the Hamiltonian, i.e., by the matrix $V=V_1V_2V_3$. Their explicit forms are

$$\tilde{\mathrm{l}}_1=\begin{vmatrix}[(\cos K+\sin K\sin\varphi)(\cos L+\sin L)+\cos\varphi(\cos L-\sin L)]/2\\ [\sin\varphi(\cos L-\sin L)-\sin K\cos\varphi(\cos L+\sin L)]/\sqrt{2}\\ [(\cos K-\sin K\sin\varphi)(\cos L+\sin L)-\cos\varphi(\cos L-\sin L)]/2\end{vmatrix},$$

$$\tilde{\mathrm{l}}_2=\begin{vmatrix}(\sin K-\cos K\sin\varphi)/\sqrt{2}\\ \cos K\cos\varphi\\ (\sin K+\sin\varphi\cos K)/\sqrt{2}\end{vmatrix}, \tag{17}$$

$$\tilde{\mathrm{l}}_3=\begin{vmatrix}[(\cos K+\sin K\sin\varphi)(\cos L-\sin L)-\cos\varphi(\cos L+\sin L)]/2\\ -[\sin\varphi(\cos L+\sin L)+\sin K\cos\varphi(\cos L-\sin L)]/\sqrt{2}\\ [(\cos K-\sin K\sin\varphi)(\cos L-\sin L)+\cos\varphi(\cos L+\sin L)]/2\end{vmatrix}.$$

Formulas (16), (17), and (14), together with (15), complete the solution of the problem of finding the eigen-

values and eigenvectors of the one-ion problem.

If we are interested only in the eigenvalues of the Hamiltonian, we can confine ourselves to a simpler program—perform only two transformations V_1 and V_2, as a result of which the Hamiltonian

$$\mathcal{H}_1(\varphi, K)=V_2V_1\mathcal{H}_1V_1^{-1}V_2^{-1}$$

takes the form

$$\mathcal{H}_1(\varphi, K)=-H(\varphi, K)\sum_i S_i^z+D_0(\varphi, K)\sum_i O_i^0+D_2(\varphi, K)\sum_i (O_i^2+O_i^{-2}), \quad (18)$$

where $D_0(\varphi,K)=d(\varphi,K)$ and $d(\varphi,K)$, $H(\varphi,K)$ and $D_2(\varphi,K)$ are defined by formula (14).

Since $[O_i^0,\mathcal{H}(\varphi,K)]=0$, the eigenvectors of the Hamiltonian are classified in accord with the eigenvectors of the operator O^0, i.e., only the states $|1\rangle$ and $|-1\rangle$ remain entangled. The eigenvalue equation therefore splits into a quadratic and a linear equation and can be easily solved:

$$E_1={}^1/_3D_0-(H^2+D_2^2)^{1/2},\ E_2=-{}^2/_3D_0,\quad E_3={}^1/_3D_0+(H^2+D_2^2)^{1/2}. \quad (19)$$

The angles φ and K are determined from the equations

$$\begin{aligned}&(\varepsilon-d)\sin\varphi\cos\varphi\cos K-(h_\parallel\cos\varphi+h_\perp\sin\varphi)\sin K=0,\\ &-[d\cos^2\varphi+\varepsilon(1+\sin^2\varphi)]\sin K\cos K+(h_\perp\cos\varphi-h_\parallel\sin\varphi)\cos 2K=0.\end{aligned} \quad (20)$$

CONCLUSION

The obtained solution of the one-ion problem can be used directly in the interpretation of the fine structures of EPR and EAR, and after simple additional operations also to determine the magnetic susceptibility, the thermodynamic function, and any other characteristic of a system with paramagnetic ions in a diamagnetic matrix. In addition, the developed approach can be used to solve, in the molecular-field approximation, the problem of the behavior of a ferromagnet (antiferromagnet) with OA of the type (1) in an arbitrarily directed magnetic field. Indeed, by making the same unitary transformations (in the simplified second variant) in $\mathcal{H}_1$ and $\mathcal{H}_{\text{int}}$, and then replacing the operators in the exchange part of the Hamiltonian by their mean values in the spirit of the molecular-field approximation (MFA), we obtain a Hamiltonian in the form (18). The formulas (14) for $H(\varphi,K)$, $D_2(\varphi,K)$ and $d(\varphi,K)$ and the Eqs. (20) for φ and K acquire then additional terms connected with the mean values characteristic of the problem, namely the order parameters. The equations for them can be easily obtained in explicit form by starting from the Hamiltonian (18), for which the calculation of the partition function and of the free energy entails no difficulty. Together with the equations for φ and K they form a complete system of self-consistent equations, which provides the solution of the problem in the MFA.

To construct the spectrum of the elementary excitations on the ground state of a ferromagnet (antiferromagnet), determined in the MFA, or else the spectrum of magnetic excitons in a paramagnet, it is necessary to use the first and most detailed variant of the transformations, since knowledge of the explicit form of the eigenvectors is essential. Inasmuch as the elementary-excitation spectrum is invariant to unitary transformations, it is convenient to carry out all the operations in a basis defined by the angles φ, K, and L, where the eigenvectors have the simplest possible form.

All the transformations were made in the present paper for $S=1$. In the case of arbitrary S, the Hamiltonian $\mathcal{H}^a$ can contain in addition to (or in lieu of) the quadratic terms also terms of higher order in the spin operators. In the general case the complete set of operators that characterize the state of the ion makes up a Lie algebra $SU(2S+1)$,[8] whose rank is equal to $2S$. This means that the one-ion Hamiltonian can be reduced to the form

$$\mathcal{H}_1=-h(\varphi_1,\varphi_2,\ldots)\sum_i S_i^z+d(\varphi_1,\varphi_2,\ldots)\sum_i O_2^0(i)$$
$$+\ldots+f(\varphi_1,\varphi_2,\ldots)\sum_i O_{2s}^0(i).$$

The unitary transformations needed for a reduction to this form are generated by the remaining operators from $ASU(2S+1)$. They can be carried out explicitly by the same method as used for the transformations of V_2 and V_3 for $S=1$.

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[1)] Here and elsewhere we omit for brevity the subscript $l=2$ of the operators O_2^m, and write, as usual, S^m in place of the operators O_1^m.

[2)] Introduction of the coefficient $2^{-1/2}$ is aimed at an exact reduction of V_1 to a rotation around the y axis.

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