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External electric field and strain effects in a quantum paramagnet

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Influence of the external electric field and the external strain on a quantum paramagnetic insulator is studied. It is shown that the external electric field and/or strain can cause drastic changes in the magnetic characteristics of the paramagnet.

Keywords: magnetic anisotropy, electric field, external strain, magnetic moment.

Magneto-electric, piezoelectric, and magneto-elastic effects are among the most studied effects in modern magnetism last years. The interest to these effects is in the potential possibility of using such effects in practice, spintronics being the prime example of such application. Also, it is possible to use systems, in which such effects can be manifested, for the storage, writing and reading of information. Mostly, such effects reveal themselves in so-called multiferroics, where both magnetic and ferroelectric ordering take place [1–5]. Mentioned effects are related to the manifestation of the strong interaction between the spin, orbital, charge, and elastic subsystems. Most of the studies were performed on magnetically ordered systems, ferro- and antiferromagnets (like ferroborates, [6–14]), see, e.g., Refs. 15–18, in which magneto-electric, magneto-elastic, and piezomagnetic properties of multiferroics were studied.

From general grounds it is obvious that similar effects can exist in magnetic systems without spin ordering, in paramagnets. Recently we have proposed the study, in which the renormalization of elastic, piezoelectric, and electric characteristics of a quantum paramagnet was theoretically calculated [19]. Similar effects were recently observed in the alumoborate of Ho [20].

The aim of the present work is to consider the effects of the external electric field or the external strain on magnetic characteristics of a quantum paramagnet due to the coupling between the electric, magnetic, and elastic subsystems of the crystal. We suppose that the external electric field E and the external strain u_0 can be considered as classical variables. Namely, we study the orthorhombic paramagnetic crystal with magnetic ions surrounded by ligands. The latter determine the crystalline electric field, which acts on magnetic ions, and, together with the spin-orbit interaction, defines the magnetic anisotropy in the effective spin model. The external electric field itself can change the effective field, which acts on magnetic ions, together with the internal crystalline electric field, and, in turn, can change the value of the parameters of the magnetic anisotropy. On the other hand, the external strain can change the positions of ligands, thus changing the value of the crystalline electric field, which acts on the magnetic ions. As a result, similarly to the above effect of the external electric field, it changes the effective values of the parameters of the magnetic anisotropy of the paramagnet. In our study, we calculate how such effects can be observed in the magnetic field dependences of the magnetic characteristics of such a crystal.

Let us first study the effect of the electric field on the magnetic characteristics of the paramagnet. Consider the Hamiltonian [19, 22]

$$\mathcal{H} = -g_{\text{eff}}\mu_B H S_z - B_1 O_0^2 - B_2 O_2^2 - \varepsilon\frac{E^2}{8\pi} + aEO_2^2 , \quad (1)$$

where g_{eff} is the effective g factor for the magnetic field H (supposed to be directed along the z axis), μ_B is the Bohr magneton, $O_0^2 = S_z^2 - S(S+1)/3$ and $O_2^2 = S_x^2 - S_y^2$ are the Stevens operators, see, e.g., [21] (operators of the components of the quadrupolar tensor), $B_{1,2}$ are the parameters of the magnetic anisotropy, $E \equiv E_x$ is the electric field directed along the x axis, ε is the electric permittivity, and a is the coefficient of the magneto-electric coupling. In Ref. 19 from the equation for the electric induction $D = -4\pi\partial F / \partial E$ and the definition of the electric permittivity $\varepsilon = \partial D / \partial E$ we found the effective electric permittivity

$$\varepsilon_{\text{eff}} = \varepsilon - 4\pi a \frac{\partial Q}{\partial E}, \tag{2}$$

where $Q = \langle O_2^2 \rangle$ (the brackets denote the averaging with the density matrix) is the average value of the component of the quadrupole operator. The value of Q depends on the external magnetic field [19], hence changing its value one can observe the magneto-electric effect on the quantum paramagnet, in which the external magnetic field affects the electric characteristic of the studied system.

Let us consider the opposite effect, how the external electric field changes the magnetic characteristics of a paramagnet. We denote $B_2^{\text{eff}} = B_2 - aE$. In particular, it follows that $Q = -\partial F / \partial B_2^{\text{eff}}$, and the in-plane component of the tensor of the quadrupolar susceptibility is $\chi_Q = \partial Q / \partial B_2^{\text{eff}}$.

In the case $S = 1$, the free energy of the system in the magnetic field directed along z axis can be written as [23]

$$F = \frac{2B_1}{3} - k_B T \ln\left[1 + 2\exp\left(\frac{B_1}{k_B T}\right)\cosh\left(\frac{A}{k_B T}\right)\right], \tag{3}$$

where k_B is the Boltzmann constant, T is the temperature, and $A = \sqrt{(B_2^{\text{eff}})^2 + (g_{\text{eff}}\mu_B H)^2}$. Then we can obtain the expressions for the studied component of the magnetic moment and the magnetic susceptibility of the paramagnet:

$$M = \frac{2g_{\text{eff}}\mu_B H \exp\left(\frac{B_1}{k_B T}\right)\sinh\left(\frac{A}{k_B T}\right)}{AZ_1}, \tag{4}$$

where $Z_1 = 1 + 2\exp(B_1 / k_B T)\cosh(A / k_B T)$, and

$$\chi = \frac{2(g_{\text{eff}}\mu_B)^2 \exp\left(\frac{B_1}{k_B T}\right)}{A^2 Z_1}\left\{\frac{(B_2^{\text{eff}})^2}{A}\sinh\left(\frac{A}{k_B T}\right) + \right.$$
$$\left. + \frac{(g_{\text{eff}}\mu_B H)^2}{k_B T Z_1}\left[\cosh\left(\frac{A}{k_B T}\right) + 2\exp\left(\frac{B_1}{k_B T}\right)\right]\right\}. \tag{5}$$

At zero magnetic field, $H = 0$, the magnetic moment is zero, naturally, while the magnetic susceptibility is nonzero:

$$\chi = \frac{2(g_{\text{eff}}\mu_B)^2 e^{B_1/k_B T}\sinh(B_2^{\text{eff}} / k_B T)}{[1 + 2e^{B_1/k_B T}\cosh(B_2^{\text{eff}} / k_B T)]}.$$

In the ground state for $B_2^{\text{eff}} \neq 0$ the magnetic moment is $M = g\mu_B H / A$, and the magnetic susceptibility is $\chi = (g_{\text{eff}}\mu_B)^2 (B_2^{\text{eff}})^2 / A^3$. It is interesting to note that for the value of the magnetic field $E > B_2 / a$ the sign of the effective parameter of the magnetic anisotropy B_2^{eff} is changed to the opposite one. That behavior is illustrated in Figs. 1 and 2, where the magnetic and the electric field dependence of the magnetic moment and the magnetic susceptibility for the quantum orthorhombic paramagnet are shown for the easy-axis-like case (notice that we consider the bi-axial case, hence, by the "easy-axis-like" and the "easy-plane-like" cases we mean the sign of B_1; the situation will be exactly easy-axis or easy-plane for $E = E_c = B_2 / a$, at which there is only O_0^2 operator of the anisotropy in the Hamiltonian) $B_1 = 3$ (arbitrary units) with $B_2 = 1$ and $k_B T = 0.1$ (we use the units in which $g_{eff}\mu_B = 1$).

Figures 3 and 4 show similar characteristics for the easy-plane-like $B_1 = -3$ spin $S = 1$ paramagnet.

It is seen that really the external magnetic field can dramatically change the magnetic field behavior of the magnetic moment and the magnetic susceptibility of the orthorhombic $S = 1$ paramagnet. At the critical electric

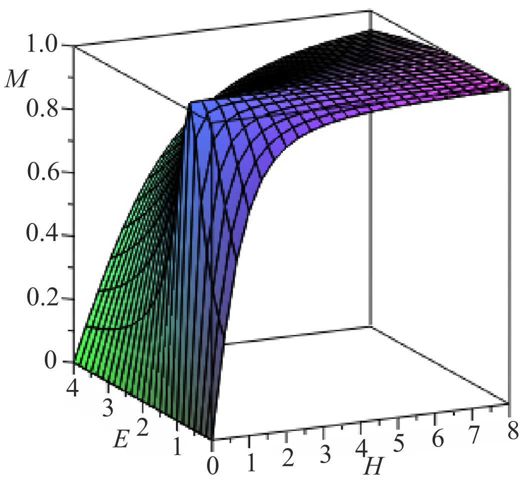

Fig. 1. (Color online) Magnetic moment of the easy-axis-like $B_1 = 3$ spin $S = 1$ paramagnet for $B_2 = 1$ and $k_B T = 0.1$ as a function of the external magnetic field H and electric field E.

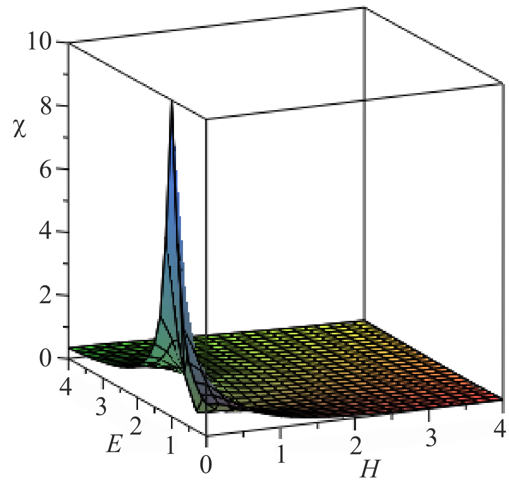

Fig. 2. (Color online) Magnetic susceptibility of the easy-axis-like $B_1 = 3$ spin $S = 1$ paramagnet for $B_2 = 1$ and $k_B T = 0.1$ as a function of the external magnetic field H and the electric field E.

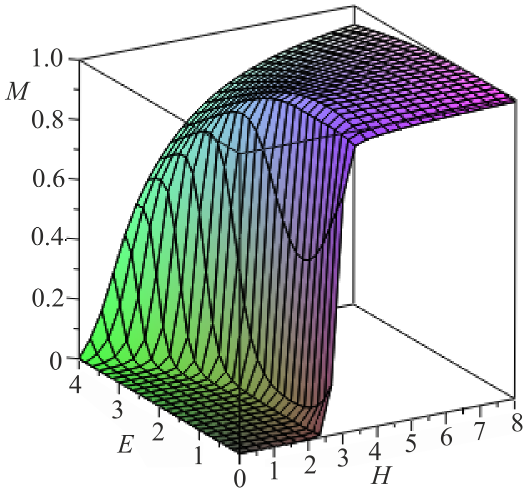

Fig. 3. (Color online) Magnetic moment of the easy-plane-like $B_1 = -3$ spin $S = 1$ paramagnet for $B_2 = 1$ and $k_BT = 0.1$ as a function of the external magnetic field H and the the electric field E.

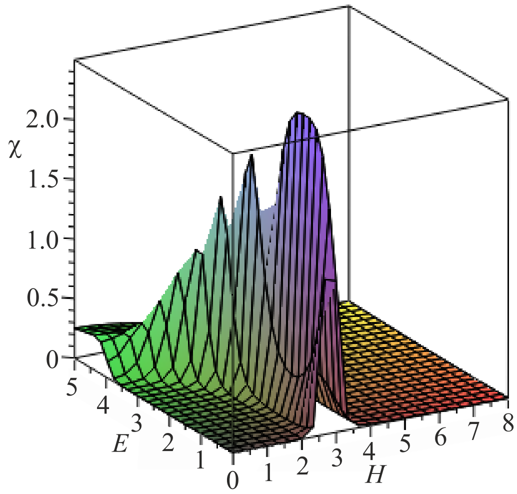

Fig. 4. (Color online) Magnetic susceptibility of the easy-plane-like $B_1 = -3$ spin $S = 1$ paramagnet for $B_2 = 1$ and $k_BT = 0.1$ as a function of the external magnetic field H and the electric field E.

field $E = E_c$ the paramagnet becomes uniaxial, and there exists two crossover values of the magnetic field, $H = 0$ for the ground state easy-axis magnet, and $H = -B_1 / g_{\text{eff}}\mu_B$ for the easy-plane magnet.

The partition function of the spin $S = 3/2$ orthorhombic quantum paramagnet in the magnetic field directed along z axis can be written as [19, 23]

$$Z = 2\left[\exp\left(-\frac{g_{\text{eff}}\mu_B H}{2k_BT}\right)\cosh\left(\frac{A_-}{k_BT}\right)+ + \exp\left(\frac{g_{\text{eff}}\mu_B H}{2k_BT}\right)\cosh\left(\frac{A_+}{k_BT}\right)\right], \quad (6)$$

where $A_\pm = \sqrt{(B_1 \pm g_{\text{eff}}\mu_B H)^2 + 3(B_2^{\text{eff}})^2}$. The magnetic moment is

$$M = \frac{g_{\text{eff}}\mu_B}{Z}\left\{\exp\left(\frac{g_{\text{eff}}\mu_B H}{2k_BT}\right)\cosh\left(\frac{A_+}{k_BT}\right)\times \left[1+\frac{2(g_{\text{eff}}\mu_B H + B_1)}{A_+}\tanh\left(\frac{A_+}{k_BT}\right)\right] - \exp\left(-\frac{g_{\text{eff}}\mu_B H}{2k_BT}\right)\cosh\left(\frac{A_-}{k_BT}\right)\times \left[1-\frac{2(g_{\text{eff}}\mu_B H - B_1)}{A_-}\tanh\left(\frac{A_-}{k_BT}\right)\right]\right\}. \quad (7)$$

The expression for the magnetic susceptibility is very long, and this is why we will not present it here. At $H = 0$, the magnetic moment is zero, while the magnetic susceptibility is nonzero:

$$\chi = \frac{(g_{\text{eff}}\mu_B)^2}{4}\left[\frac{1}{k_BT}\left(1+\frac{4B_1^2}{A_0^2}\right)+ + \frac{4}{A_0}\tanh\left(\frac{A_0}{k_BT}\right)\left(1-\frac{4B_1^2}{A_0^2}+\frac{B_1}{k_BT}\right)\right], \quad (8)$$

where $A_0 = \sqrt{B_1^2 + 3(B_2^{\text{eff}})^2}$.

Figures 5 and 6 show the magnetic and the electric field dependences of the magnetic moment and the magnetic susceptibility for the ease-axis-like ($B_1 = 3$) spin $S = 3/2$ for $B_2 = 1$ and $k_BT = 0.1$.

Figures 7 and 8 show the magnetic and the electric field dependences of the magnetic susceptibility of the orthorhombic easy-plane-like $B_1 = -3$ paramagnet for the fixed value of the in-plane anisotropy $B_2 = 1$.

It is seen that the external electric field for higher-spin paramagnet also can drastically change the behavior of the magnetic characteristics of the system. At the critical value of the electric field E_c, the magnetic susceptibility manifests crossovers in the ground state. For the easy-axis paramagnet, the crossover takes place at $H = 0$, while for the easy-plane paramagnet it happens at $H = 0$ and $H = -B_1 / g_{\text{eff}}\mu_B$.

Unfortunately, it is impossible to obtain analytic results for the magnetic field directed along other axes of the crystal for the considered Hamiltonian. However, for a slightly modified Hamiltonian it is possible to manage [24]. Namely, consider the Hamiltonian

$$\mathcal{H}_b = -g_p\mu_B H_x S_x - B_1 S_z^2 - B_2 S_x^2 - \varepsilon\frac{E^2}{8\pi} + aES_x^2, \quad (9)$$

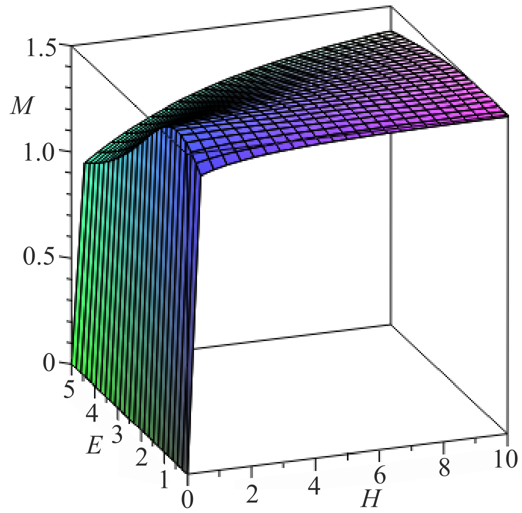

Fig. 5. (Color online) Magnetic moment of the easy-axis-like $B_1 = 3$ spin $S = 3/2$ paramagnet for $B_2 = 1$ and $k_B T = 0.1$ as a function of the external magnetic field H and the electric field E.

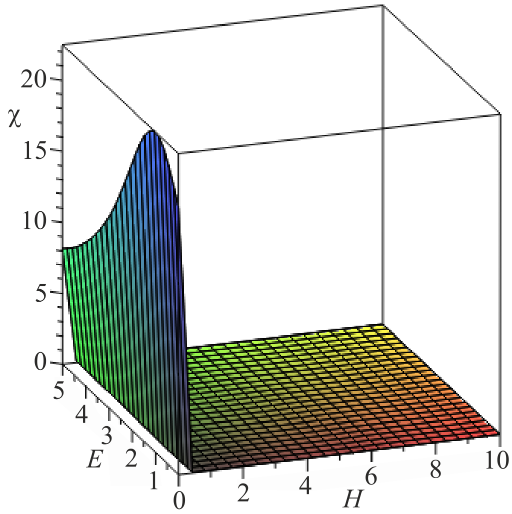

Fig. 6. (Color online) Magnetic susceptibility of the easy-axis-like $B_1 = 3$ spin $S = 3/2$ paramagnet for $B_2 = 1$ and $k_B T = 0.1$ as a function of the external magnetic field H and the electric field E.

where g_p is the effective g factor for the magnetic field H_x directed along the x axis. Performing similar calculations, we obtain, for example, for the magnetic moment for $S = 1$

$$M_x = \frac{2g_p\mu_B H_x \exp\left(\dfrac{-2B_1 + B_2^{\text{eff}}}{2k_B T}\right)\sinh\left(\dfrac{A_p}{k_B T}\right)}{A_p Z_{1p}}, \quad (10)$$

where

$$A_p = [(2g_p\mu_B H_x)^2 + B_1^2]^{1/2}/2,$$

and $Z_{1p} = 1 + \exp[(-2B_1 + B_2^{\text{eff}})/2k_B T]\cosh(A_p/k_B T)$. For $S = 3/2$ paramagnet, we get

$$M = \frac{g_p\mu_B}{Z_p}\left\{\exp\left(\frac{g_p\mu_B H_x}{2k_B T}\right)\cosh\left(\frac{A_{p+}}{k_B T}\right)\times\right.$$
$$\times\left[1 + \frac{g_p\mu_B H_x + B_2^{\text{eff}} - B_1/2}{A_{p+}}\tanh\left(\frac{A_{p+}}{k_B T}\right)\right] -$$
$$-\exp\left(-\frac{g_p\mu_B H_x}{2k_B T}\right)\cosh\left(\frac{A_{p-}}{k_B T}\right)\times$$
$$\left.\times\left[1 - \frac{g_p\mu_B H_x - B_2^{\text{eff}} + B_1/2}{A_{p-}}\tanh\left(\frac{A_{p-}}{k_B T}\right)\right]\right\}, \quad (11)$$

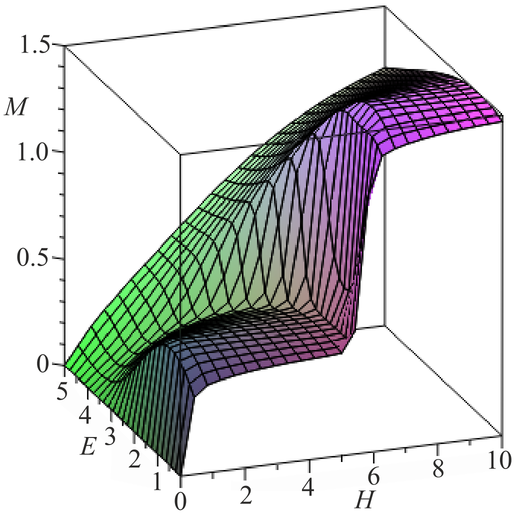

Fig. 7. (Color online) Magnetic moment of the easy-plane-like $B_1 = -3$ spin $S = 3/2$ paramagnet for $B_2 = 1$ and $k_B T = 0.1$ as a function of the external magnetic field H and the electric field E.

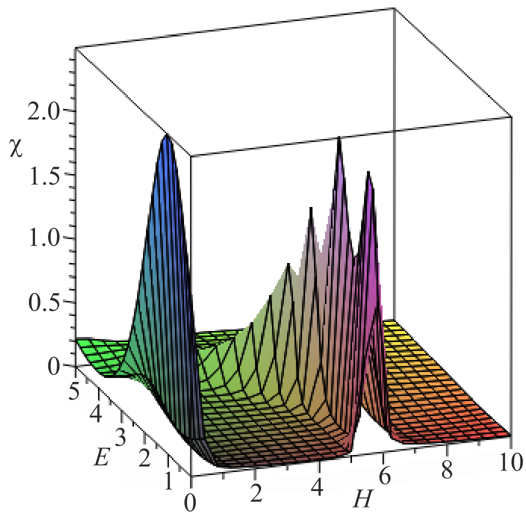

Fig. 8. (Color online) Magnetic susceptibility of the easy-plane-like $B_1 = -3$ spin $S = 3/2$ paramagnet for $B_2 = 1$ and $k_B T = 0.1$ as a function of the external magnetic field H and the electric field E.

where $A_{p\pm} = [3B_1^2 + (2B_2^{eff} - B_1 \pm 2g_p\mu_B H_x)^2]^{1/2}/2$ and

$$Z_p = 2\left[\exp\left(-\frac{g_p\mu_B H_x}{2k_BT}\right)\cosh\left(\frac{A_{p-}}{k_BT}\right) + \right.$$
$$\left. + \exp\left(\frac{g_p\mu_B H_x}{2k_BT}\right)\cosh\left(\frac{A_{p+}}{k_BT}\right)\right]. \quad (12)$$

In Figures 9 and 10, the behavior of the magnetic moment of such a paramagnet as a function of the external fields E and H_x are shown for the more interesting case $B_1 > 0$, namely for $B_1 = 3$, $B_2 = 1$, $a = 1$, $g_p\mu_B = 1$ and $k_BT = 0.1$ for $S = 1$ and $S = 3/2$. In fact, it is seen that the effect of the external electric field for such a geometry of the magnetic field is not dramatic. It only renormalizes the effective value of the magnetic anisotropy B_2. On the other hand, for the model with the Hamiltonian $\mathcal{H}_b$, one can also calculate the magnetic moment and the magnetic susceptibility for the magnetic field directed along z axis also [24]. The easiest way is to replace $B_1 \to B_2^{\text{eff}}$ and vice versa, with $g_p\mu_B H_x \to g_{\text{eff}}\mu_B H$ in the above expressions. Then it is easy to see that the drastic effect of the external electric field takes place: There is a critical value of the electric field E_c at which the system becomes uniaxial, and critical crossover phenomena take place as in the studied before more realistic system.

Now let us turn to the study of the effect of the external strain on the magnetic characteristics of the paramagnet. The Hamiltonian of the considered model can be written as [19, 25]

$$\mathcal{H} = -g_{\text{eff}}\mu_B HS_z - B_1O_0^2 - B_2O_2^2 + \frac{Cu^2}{2} -$$
$$-\varepsilon\frac{E^2}{8\pi} + eEu + aEO_2^2 + b(u-u^0)O_2^2, \quad (13)$$

where e is the piezoelectric modulus, C is the elastic modulus, u is the strain, and b is the coefficients of the magneto-elastic coupling (all issues are connected with the coordinate x). We see that the external strain u_0 plays similar to the external electric field E role: It renormalizes the effective parameter of the cristalline electric field B_2. The effect of the external strain is similar to the effect of the external electric field. However, the realistic values of the external electric field can be much larger than the one for the external strain, caused by the external pressure. This is why, it is possible that for realistic values of the external pressure one cannot reach the values of u_0 at which the crossover to the uniaxial paramagnet can take place. Notice that the following relation for the strain holds:

$$\frac{\partial^2 u}{\partial t^2} - \left[C - \frac{be}{a}\right]\frac{\partial u}{\partial x} = \left[e + \frac{b\varepsilon}{4\pi a}\right]\frac{\partial E}{\partial x}, \quad (14)$$

i.e., the dynamics of the strain in the system depends on the spatial changes of the external electric field.

We point out that the effects, discussed above, can be observed easier for rare-earth based paramagnets, rather than for transition metal-based, because in the former the crystalline electric field is weaker, and the critical values of the external electric field and strain can be achieved easier.

In summary, we have shown that the external electric field and the external strain can change the magnetic field behavior of magnetic characteristics of a quantum paramagnet. In particular, we have shown that the most drastic changes take place if the electrtic field is directed perpendicular to the direction of the magnetic field. In such a situation, the external electric field can change drastically the magnetic field dependences of the magnetic moment and susceptibility. On the other hand, for the parallel orientation of the external electric and magnetic fields,

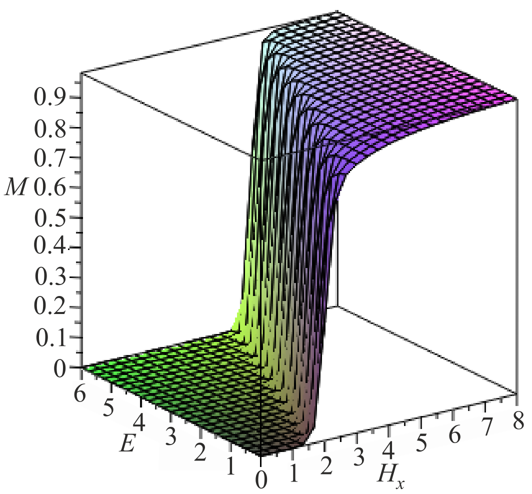

Fig. 9. (Color online) Magnetic moment of the $B_1 = 3$ spin $S = 1$ biaxial paramagnet for $B_2 = 1$ and $k_BT = 0.1$ as a function of the external magnetic field H_x and the electric field E.

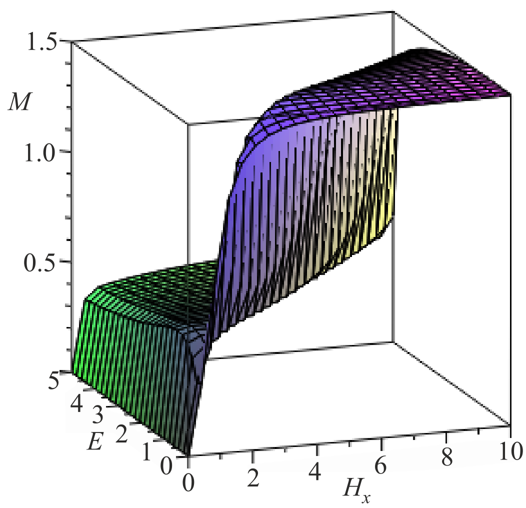

Fig. 10. (Color online) Magnetic moment of the $B_1 = 3$ spin $S = 3/2$ biaxial paramagnet for $B_2 = 1$ and $k_BT = 0.1$ as a function of the external magnetic field H_x and the electric field E.

the changes in the behavior also take place, however, they are rather quantitative. We suppose that the predicted effects can be observed in rare-earth based paramagnetic insulators, with relatively small orthorhombic anisotropy.

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Ефекти зовнішнього електричного поля та деформації в квантовому парамагнетику

A. A. Zvyagin

Вивчено вплив зовнішнього електричного поля та зовнішньої деформації на квантовий парамагнітний діелектрик. Показано, що зовнішне електричне поле та/або деформація можуть привести до суттєвих змін магнітних характеристик парамагнетика в магнітному полі.

Ключові слова: магнітна анізотропія, електричне поле, зовнішня деформація, магнітний момент.