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Piezoelectric and magneto-elastic effects in a quantum paramagnet

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Electric, piezoelectric, and elastic characteristics of the orthorhombic quantum paramagnet are calculated. Using the exact analytical solution it was shown that the electric permittivity, piezoelectric, and elastic modules can manifest strong dependences on the values of the external magnetic, electric field, external strain, and temperature.

Keywords: electric permittivity, elastic modulus, piezoelectric modulus, magnetic anisotropy.

Last years magneto-electric, piezoelectric, and magneto-elastic effects attract the attention of researchers. Those effects are the manifestation of the coupling between the electric, magnetic, and elastic subsystems of the studied compounds. The interest to the studies of such effects is stimulated by the possibilities to utilize them in dissipation-free systems for writing and reading an information. The most attractive subjects for such a purpose are so-called multiferroics, which have both magnetic and ferroelectric properties (see, e.g., [1–4]). The magneto-electric response appears there due to the action of the own exchange magnetic field, and used in experiments the external magnetic field is applied mostly for the monodomenization of investigated samples. This is why, most of studies were performed on magnetically ordered systems, ferro- and antiferromagnets (like ferroborates, see, e.g., [5–7]), for the recent studies see [8–10]. However, it is clear from general grounds that similar effects can exist in magnetic systems without spin ordering, i.e., in quantum paramagnets.

The goal of the present study is to find the effects of the renormalization of electric, piezoelectric, and elastic characteristics of a paramagnet due to the coupling between the electric, magnetic, and elastic subsystems of the crystal. Namely, we investigate 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. Then the interaction between the spin, charge and elastic subsystems of the crystal can yield magneto-electric, piezoelectric, and magneto-elastic effects in such a quantum paramagnet. Below we calculate how such an interaction can be observed in the temperature and magnetic field dependences of the characteristics of such a crystal, like the electric permittivity, piezoelectric, and elastic modules.

The Hamiltonian of the considered model can be written as

$$\mathcal{H} = -g_{\text{eff}}\mu_B H S_z - B_1 O_0^2 - B_2 O_2^2 + \frac{Cu^2}{2} -$$
$$-\varepsilon\frac{E^2}{8\pi} + eEu + aEO_2^2 + b(u-u_0)O_2^2 , \qquad (1)$$

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., [11] (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, e is the piezoelectric modulus, C is the elastic modulus, u is the strain (u_0 is the static strain), and a and b are the coefficients of the magneto-electric and magneto-elastic couplings, respectively (all issues are connected with the coordinate x). Using the standard formulas of the elasticity theory

$$\sigma = \frac{\partial F}{\partial u} = Cu + eE + bQ , \qquad (2)$$

where F is the free energy of the system, σ is the elastic deformation, $Q = \langle O_2^2 \rangle$ (the brackets denote the averaging with the density matrix) is the average value of the component of the quadrupole operator, and the definition

$$e = \frac{d\sigma}{dE}, \qquad (3)$$

for the effective piezoelectric modulus [12], we get

$$e_{\text{eff}} = e + b\frac{\partial Q}{\partial E}. \tag{4}$$

Then using the equation for the electric induction D

$$D = -4\pi\frac{\partial F}{\partial E} \tag{5}$$

and the definition of the electric permittivity

$$\varepsilon = \frac{\partial D}{\partial E} \tag{6}$$

one can find the effective permittivity

$$\varepsilon_{\text{eff}} = \varepsilon - 4\pi a\frac{\partial Q}{\partial E}. \tag{7}$$

Finally, according to the elasticity theory [13] we have

$$\rho\frac{\partial^2 u}{\partial t^2} = \frac{d\sigma}{dx}, \tag{8}$$

where ρ is the density of the crystal. Calculating the right hand side of that equation we get

$$\frac{\partial\sigma}{\partial x} = C\frac{\partial u}{\partial x} + e\frac{\partial E}{\partial x} + b\left(\frac{\partial Q}{\partial u}\frac{\partial u}{\partial x} + \frac{\partial Q}{\partial E}\frac{\partial E}{\partial x}\right). \tag{9}$$

Then, using the equation of the electric neutrality (we use here only the necessary component of the electric induction)

$$(\nabla\cdot D) = \varepsilon\frac{\partial E}{\partial x} - 4\pi e\frac{\partial u}{\partial x} - 4\pi a\left(\frac{\partial Q}{\partial u}\frac{\partial u}{\partial x} + \frac{\partial Q}{\partial E}\frac{\partial E}{\partial x}\right) = 0 \tag{10}$$

we obtain

$$\frac{\partial\sigma}{\partial x} = \left(\frac{\partial u}{\partial x}\right)\left[C + 4\pi e\frac{e_{\text{eff}}}{\varepsilon_{\text{eff}}} + \left(b + 4\pi a\frac{e_{\text{eff}}}{\varepsilon_{\text{eff}}}\right)\frac{\partial Q}{\partial u}\right]. \tag{11}$$

The right hand side of the latter can be presented via the effective elastic modulus C_{eff}. Let us denote $B_2^{\text{eff}} = B_2 - aE - b(u - u_0)$. We obtain $Q = -\partial F/\partial B_2^{\text{eff}}$, and $\partial Q/\partial E = -a(\partial Q/\partial B_2^{\text{eff}})$ and $\partial Q/\partial u = -b(\partial Q/\partial B_{\text{eff}})$. It can be taken into account that $\partial Q/\partial B_2^{\text{eff}} = \chi_Q$ is the in-plane component of the tensor of the quadrupolar susceptibility. Then it is clear that all above mentioned characteristics of the system can be presented as a function of that component of the quadrupolar susceptibility. Below, we present analytical results for that component of the quadrupolar susceptibility as a function of the temperature, applied electro-magnetic field, and the parameters of the magnetic anisotropy for the orthorhombic quantum paramagnet for the cases $S = 1$ and $S = 3/2$.

The free energy of the spin $S = 1$ orthorhombic quantum paramagnet in the magnetic field directed along z axis can be written, see, e.g., [14], as (here we suppose that the electric field E and the strain u are classical)

$$F = \frac{2B_1}{3} - T\ln\left[1 + 2\exp\left(\frac{B_1}{k_B T}\right)\cosh\left(\frac{A}{k_B T}\right)\right], \tag{12}$$

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 necessary components of the quadrupolar moment and quadrupolar susceptibility

$$Q = \frac{2B_2^{\text{eff}}\exp\left(\frac{B_1}{k_B T}\right)\sinh\left(\frac{A}{k_B T}\right)}{AZ_1}, \tag{13}$$

and

$$\chi_Q = \frac{2\exp\left(\frac{B_1}{k_B T}\right)\sinh\left(\frac{A}{k_B T}\right)}{AZ_1}\times$$

$$\times\left[\frac{(g_{\text{eff}}\mu_B H)^2}{A^2} + \frac{(B_2^{\text{eff}})^2\coth\left(\frac{A}{k_B T}\right)}{k_B TA} - \right.$$

$$\left. - \frac{2(B_2^{\text{eff}})^2\exp\left(\frac{B_1}{k_B T}\right)\sinh\left(\frac{A}{k_B T}\right)}{k_B TAZ_1}\right], \tag{14}$$

where $Z_1 = 1 + 2\exp(B_1/k_B T)\cosh(A/k_B T)$. It is seen that for $B_2^{\text{eff}} = 0$ the quadrupole moment is zero, $Q = 0$, while the susceptibility

$$\chi_Q = 2\exp(B_1/k_B T)\sinh(g_{\text{eff}}\mu_B H/k_B T)/(g_{\text{eff}}\mu_B H\times$$

$$\times[1 + 2\exp(B_1/k_B T)\cosh(g_{\text{eff}}\mu_B H/k_B T)]$$

is finite, as it must be. In the ground state we have $Q = B_2^{\text{eff}}/A$ and $\chi_Q = (g_{\text{eff}}\mu_B H)^2/A^3$ (which implies zero ground-state quadrupolar susceptibility without the application of the external magnetic field). On the other hand, at high temperatures we obtain $Q \approx B_2^{\text{eff}}/k_B T$ and $\chi_Q \approx (2/3k_B T)$. In the absence of the external magnetic field, $H = 0$, one can replace $A \to B_2^{\text{eff}}$.

For the partition function of the spin $S = 3/2$ orthorhombic quantum paramagnet in the magnetic field directed along z axis we can write [14]

$$Z = 2\left[\exp\left(-\frac{g_{\text{eff}}\mu_B H}{2k_B T}\right)\cosh\left(\frac{A_-}{k_B T}\right) + \right.$$

$$\left. + \exp\left(\frac{g_{\text{eff}}\mu_B H}{2k_B T}\right)\cosh\left(\frac{A_+}{k_B T}\right)\right], \tag{15}$$

where $A_{\pm} = \sqrt{(B_1 \pm g_{\rm eff}\mu_B H)^2 + 3(B_2^{\rm eff})^2}$. The necessary components of the quadrupolar moment and the quadrupolar susceptibility are then

$$Q = \frac{6B_2^{\rm eff}}{Z}\left[\frac{\exp\left(-\frac{g_{\rm eff}\mu_B H}{2k_BT}\right)\sinh\left(\frac{A_-}{k_BT}\right)}{A_-} + \frac{\exp\left(\frac{g_{\rm eff}\mu_B H}{2k_BT}\right)\sinh\left(\frac{A_+}{k_BT}\right)}{A_+}\right], \tag{16}$$

and

$$\chi_Q = \frac{6}{Z}\left[\frac{\exp\left(-\frac{g_{\rm eff}\mu_B H}{2k_BT}\right)\sinh\left(\frac{A_-}{k_BT}\right)}{A_-}\times \left[\frac{(B_1 - g_{\rm eff}\mu_B H)^2}{A_1^2} + \frac{3(B_2^{\rm eff})^2\coth\left(\frac{A-}{k_BT}\right)}{k_BTA_-}\right] + \frac{\exp\left(\frac{g_{\rm eff}\mu_B H}{2k_BT}\right)\sinh\left(\frac{A_+}{k_BT}\right)}{A_+}\times \left(\frac{(B_1 + g_{\rm eff}\mu_B H)^2}{A_+^2} + \frac{3(B_2^{\rm eff})^2\coth\left(\frac{A_+}{k_BT}\right)}{k_BTA_+}\right)\right] - \frac{36(B_2^{\rm eff})^2}{Z^2}\left[\frac{\exp\left(-\frac{g_{\rm eff}\mu_B H}{2k_BT}\right)\sinh\left(\frac{A_-}{k_BT}\right)}{A_-} + \frac{\exp\left(\frac{g_{\rm eff}\mu_B H}{2k_BT}\right)\sinh\left(\frac{A_+}{k_BT}\right)}{A_+}\right]^2. \tag{17}$$

Again at $B_2^{\rm eff} = 0$ we have $Q = 0$ and finite quadrupole susceptibility. In the ground state we have $Q = 6B_2^{\rm eff}[A_-^{-1} + A_+^{-1}]$ and $\chi_Q = 6[A_+^2A_-^2(A_+ + A_-) - 3(B_2^{\rm eff})^2(A_+^3 + A_-^3)]/A_+^3A_-^3$. Notice that unlike the case $S = 1$ the ground-state quadrupole susceptibility is finite even at $H = 0$. At high temperatures, we obtain $Q \approx 3B_2^{\rm eff}/2k_BT$ and $\chi_Q \approx 3/2k_BT$. In the absence of the external magnetic field (which lifts the degeneracy between two doublets) we have $A_+ = A_- = \sqrt{B_1^2 + 3(B_2^{\rm eff})^2}$.

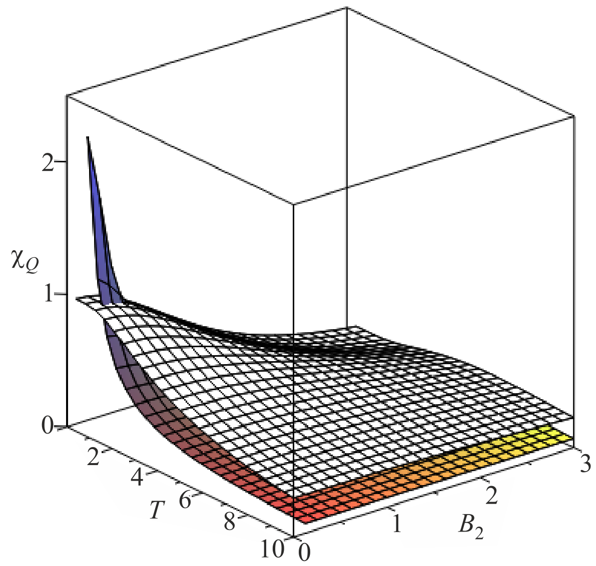

Fig. 1. (Color online) Quadrupolar in-plane susceptibility of the easy-axis $B_1 > 0$ paramagnet in the absence of external fields and the static strain as a function of the temperature and the parameter of the in-plane orthorhombic anisotropy B_2. Color surface: $S = 1$; grey surface: $S = 3/2$.

Figures 1 and 2 show the temperature and B_2-dependences of the quadrupolar susceptibility for the easy-axis ($B_1 = 3$) and the easy-plane ($B_1 = -3$) magnetic anisotropy for the absence of the extenal electric and magnetic fields $H = E = 0$ and the static strain $u_0 = 0$. From the above

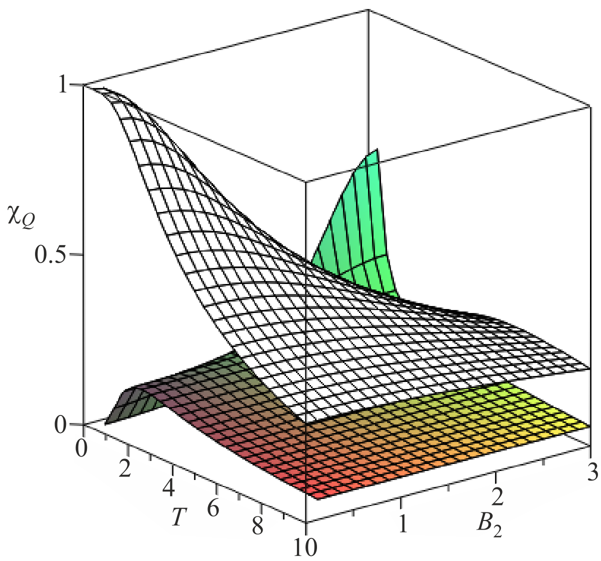

Fig. 2. (Color online) The same as in Fig. 1 but for the easy-plane $B_1 < 0$ magnetic anisotropy.

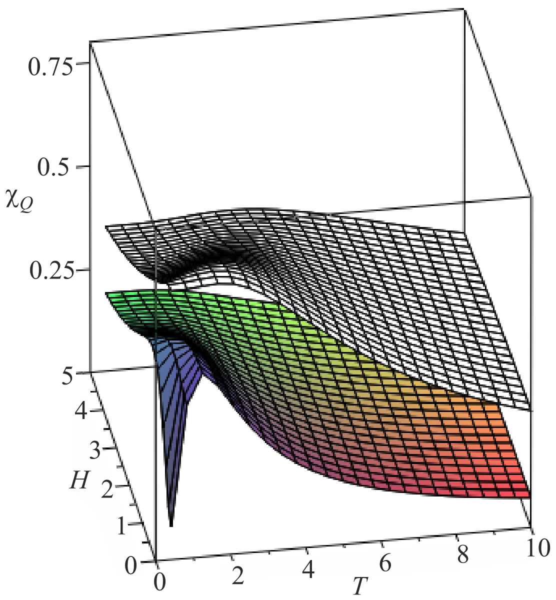

Fig. 3. (Color online) Quadrupolar in-plane susceptibility of the easy-axis paramagnet for $E = u_0 = 0$ and $B_2 = 1$ as a function of the temperature and the external magnetic field. Color surface: $S = 1$; grey surface: $S = 3/2$.

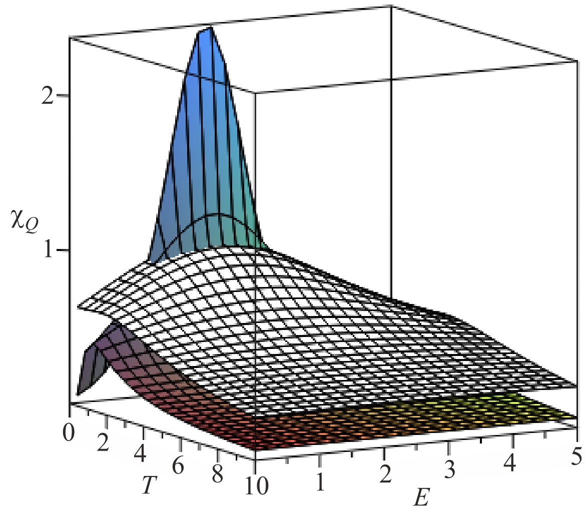

Fig. 5. (Color online) Quadrupolar in-plane susceptibility of the easy-axis paramagnet for $H = u_0 = 0$ and $B_2 = 1$ as a function of the temperature and the external electric field. Color surface: $S = 1$; grey surface: $S = 3/2$.

formulas we know that the dependences have to be the same for negative and positive B_2 for $E = u_0 = 0$. We see that the quadrupolar susceptibility, as a rule, decays at high enough temperatures, while for low temperatures it can manifest non-monotonic temperature dependence.

Figures 3 and 4 show the temperature and magnetic field dependences of the quadrupolar in-plane susceptibility of the orthorhombic paramagnet for the fixed value of the in-plane anisotropy in the absence of the external electric field and static strain. As a rule, the external magnetic field can essentially change the behavior of the quadrupolar susceptibility, especially at low temperatures.

Figures 5 and 6 show the temperature and electric field dependences of the quadrupolar in-plane susceptibility of the easy-axis and the easy-plane B_1 magnetic anisotropy, respectively, for the fixed value of the in-plane anisotropy in the absence of the external magnetic field and the static strain for $a = 0.5$. At low temperatures the electric field dependence of the quadrupolar in-plane susceptibility of the paramagnet is non-monotonic, and the position of the maximum is dependent on the value of B_2.

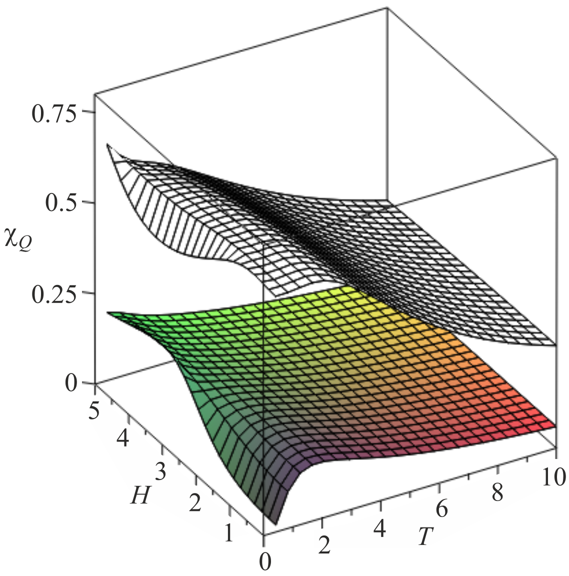

Fig. 4. (Color online) The same as in Fig. 3 but for the easy-plane B_1 magnetic anisotropy.

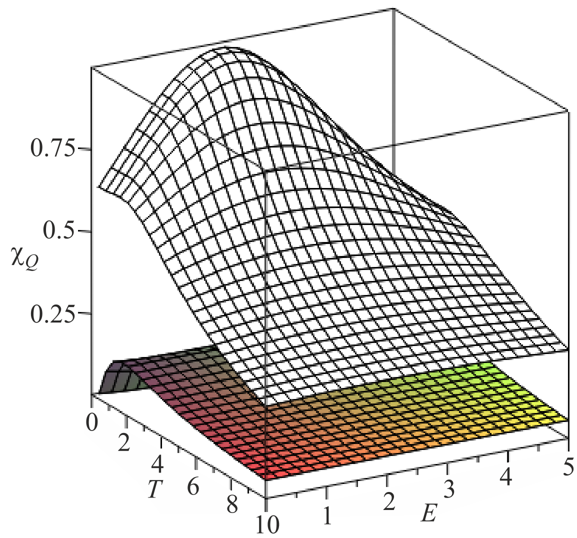

Fig. 6. (Color online) The same as in Fig. 5 but for the easy-plane magnetic anisotropy.

Then, using the above obtained results, we calculate the renormalized characteristics

$$\Delta e \equiv e_{\text{eff}} - e = -ab\chi_Q,$$

$$\Delta\varepsilon \equiv \varepsilon_{\text{eff}} - \varepsilon = 4\pi a^2\chi_Q \tag{18}$$

and

$$\Delta C \equiv C_{\text{eff}} - C = -b^2\chi_Q + 4\pi\frac{e_{\text{eff}}^2}{\varepsilon_{\text{eff}}} =$$

$$= C + \frac{4\pi e^2 - b(8\pi ae + b\varepsilon)\chi_Q}{\varepsilon + 4\pi a^2\chi_Q}. \tag{19}$$

Taking into account that the component of the quadrupole susceptibility is positive, we see that the change of the electrical permittivity $\varepsilon_{\text{eff}} - \varepsilon$ is positive. The change of the elastic modulus $C_{\text{eff}} - C$ exists even for $a = 0$, i.e., for the absent magneto-electric coupling (in that case, however, the piezoelectric coupling produces the constant term). The change obviously consists of two contributions (see the first line of the above formula). The first contribution (the magneto-elastic contribution) is negative; it manifests the softening of the elastic modulus due to the magneto-elasticity. The second term is positive; it manifests the hardening of the elastic modulus, caused by the piezoelectricity. Hence, the softening or the hardening of the elastic modulus (the sign of the change) of the considered system depends on whether the quadrupole susceptibility is larger or smaller than $4\pi e^2 / b(8\pi ae - b\varepsilon)$, see the second line. Finally, the sign of the change of the piezoelectric modulus $e_{\text{eff}} - e$ depends on the sign of $(-ab)$.

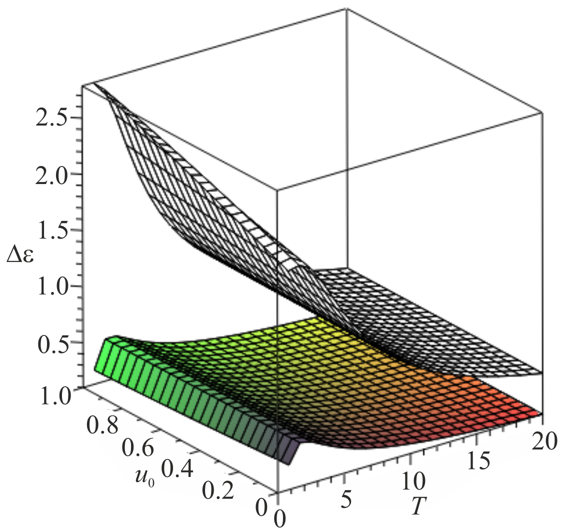

Fig. 7. (Color online) The renormalized electric permittivity of the easy-plane paramagnet $B_1 = -3$ for $B_2 = 1$ in the absence of external electric and magnetic fields $E = H = 0$ as a function of the temperature and the static strain u_0. Color surface: $S = 1$; grey surface: $S = 3/2$.

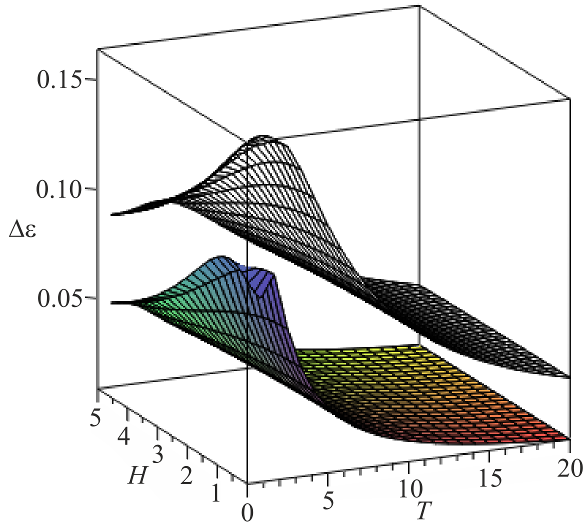

Fig. 8. (Color online) The renormalized electric permittivity of the easy-axis paramagnet $B_1 = 3$ for $B_2 = 1$ in the absence of the external electric field $E = 0$ and zero static strain $u_0 = 0$ as a function of the temperature and the external magnetic field H. Color surface: $S = 1$; grey surface: $S = 3/2$.

In Fig. 7 we present the results for the static strain dependence of the electric permittivity of the easy-plane paramagnet in the absence of the external electro-magnetic field for $a = -0.5$ and $b = 0.5$. For high enough temperatures the electric permittivity decays with T, while for small temperatures the dependence is non-monotonic. Also we see that the electric permittivity decays with the growth of the static strain u_0.

In Fig. 8 The renormalization of the electric permittivity of the easy-axis case is shown as a function of the temperature and the magnetic field for $E = u_0 = 0$.

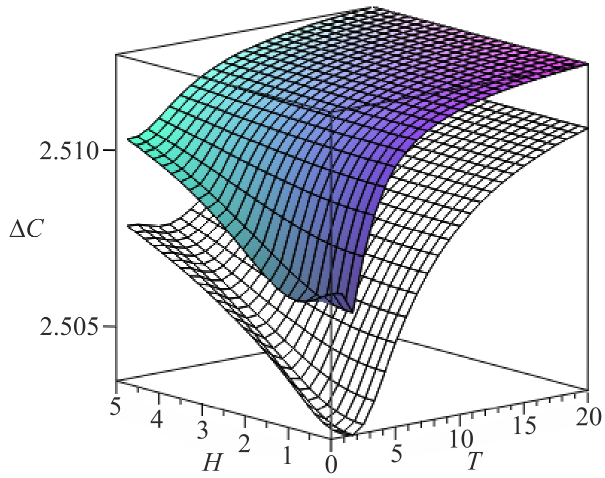

Fig. 9. (Color online) The renormalized elastic modulus of the easy-axis paramagnet $B_1 = 3$ for $B_2 = 1$ in the absence of external magnetic field and static strain $H = u_0 = 0$ as a function of the temperature and the external electric field E. Color surface: $S = 1$; grey surface: $S = 3/2$.

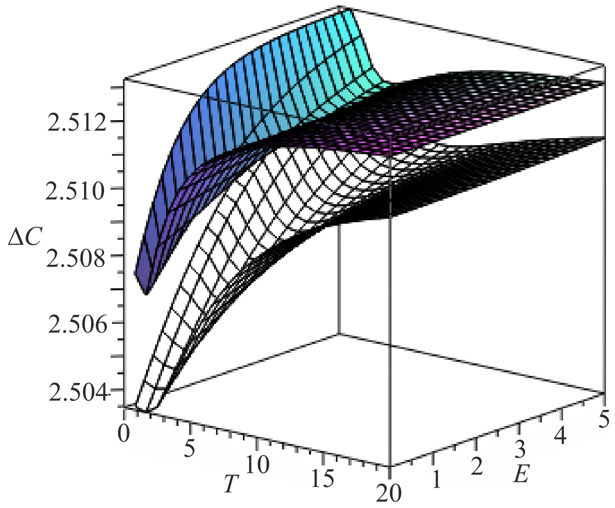

Fig. 10. (Color online) The renormalized elastic modulus of the easy-plane paramagnet $B_1 = -3$ for $B_2 = 1$ in the absence of the external magnetic field and static strain $H = u_0 = 0$ as a function of the temperature and the electric field E. Color surface: $S = 1$; grey surface: $S = 3/2$.

In Figs. 9 and 10 the renormalization of the elastic modulus of the considered paramagnet are shown as a function of the temperature and the external magnetic field, and temperature and the external electric field, respectively. We used the parameters $\varepsilon = 20$, $e = 2$, $a = -0.5$, $b = 0.5$. Both figures show the low-temperature minimum, characteristic for that kind of systems. The growth of the external electric and magnetic field induces hardening of the elastic modulus.

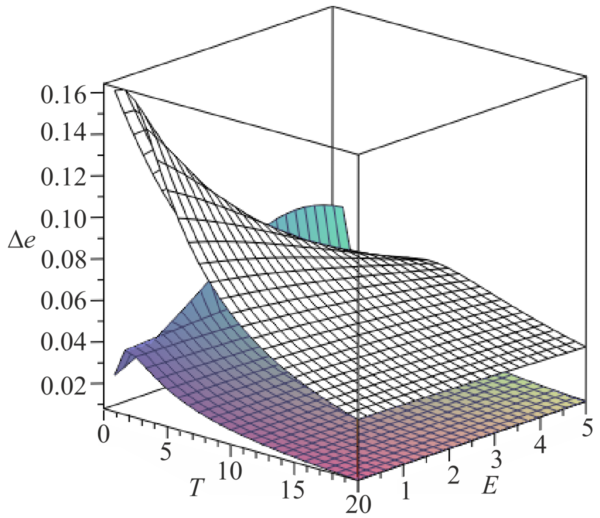

Fig. 11. (Color online) The renormalized piezoelectric modulus of the easy-plane paramagnet $B_1 = -3$ for $B_2 = 1$ in the absence of external magnetic field and the external strain $H = u_0 = 0$ as a function of the temperature and the electric field E. Color surface: $S = 1$; grey surface: $S = 3/2$.

Finally, in Fig. 11 the renormalization of the piezoelectric modulus is shown as a function of the temperature and the electric field for $H = u_0 = 0$. We see that depending on the value of spin the low-temperature piezoelectric modulus can decay or grow with the growth of the electric field.

In summary, electric, piezoelectric, and elastic characteristics of the orthorhombic quantum paramagnet have been calculated analytically. It is shown that electric permittivity, piezoelectric, and elastic modules can manifest strong dependences on the values of the external magnetic, electric field, external static strain, and temperature. We point out that the dependences of the electric permittivity, piezoelectric, and elastic modules in a quantum paramagnet were recently observed [15].

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П’єзоелектричний та магнітопружний ефекти у квантовому парамагнетику

A. A. Zvyagin, G. A. Zvyagina

Розраховано електричні, п’єзоелектричні та пружні характеристики квантового орторомбічного парамагнетика. Використовуючи точне аналитичне рішення, показано, що електрична проникність, п’єзомодуль та модуль пружності можуть виявляти сильні залежності від значень зовнішніх магнітного та електричного полів, зовнішньої пружньої напруги та температури.

Ключові слова: електрична проникність, модуль пружності, п’єзомодуль, магнітна анізотропія.