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Phase transitions in a compressible antiferromagnet with biquadratic coupling

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A two-sublattice compressible antiferromagnetic model is studied within a variational approach based on Bogoliubov's inequality and through spin-wave analysis. Our effective spin Hamiltonian contains a biquadratic exchange term, and the exchange couplings depend on external forces. We determine the complete phase diagram in the magnetic field versus absolute temperature plane for different values of the external forces. We also show that the canted-paramagnetic phase boundary can be of first or second order depending on the values of the single-ion uniaxial anisotropy and of the biquadratic exchange parameter. The evolution of the bicritical point as a function of the biquadratic term is the same as for the single-ion term.

I. INTRODUCTION

In this paper we study the phase diagram in the plane of H (applied magnetic field) versus T (absolute temperature) for a compressible antiferromagnetic model. From the experimental point of view some recent experiments have been performed on compressible antiferromagnets[1,2] where magnetoelastic effects were observed. On the other hand, some calculations performed on compressible magnetic systems focused attention on Ising models[3,4] and on $S=\frac{1}{2}$ antiferromagnets[5] where biquadratic coupling does not appear. These theoretical works have been performed within the mean-field approximation.

We consider in this work a compressible antiferromagnetic model of spins $S=1$ in an applied field directed along the axis of anisotropy. The spins are arranged in two interpenetrating simple-cubic sublattices. We take the simple magnetoelastic model of Baker and Essam[6] where the shear forces are disregarded. In this way we derive an effective spin Hamiltonian that contains, besides the force-dependent exchange couplings, a term that reveals the biquadratic exchange coupling. Using a variational approach based on Bogoliubov's inequality[7] we can determine a magnetic phase diagram for every value of the external force. We show that, depending on the value of the biquadratic parameter the spin-flop–to–paramagnetic transition can be of first or second order. We also exhibit a diagram of the biquadratic exchange versus the single-ion uniaxial anisotropy which shows the regions of first- and second-order spin-flop–paramagnetic transitions.

In the low-temperature region we determine analytic asymptotic expressions for the spin-flop–paramagnetic boundary. In this case our effective spin Hamiltonian is considered within the spin-wave theory.

Our paper is organized as follows: In Sec. II we present our compressible antiferromagnetic model and the calculations to obtain a variational free energy. In Sec. III, we exhibit the results obtained at $T=0$ and the complete phase diagram as a function of the external pressures. In Sec. IV, we obtain the spin-flop–paramagnetic phase boundary in the low-temperature region, and finally, in Sec. V, we discuss the main results obtained in this paper.

II. COMPRESSIBLE ANTIFERROMAGNETIC MODEL—VARIATIONAL FREE ENERGY

Our compressible antiferromagnetic model may be described by the following Hamiltonian:

$$\mathcal{H}=\sum_{i=1}^{N}\frac{\mathbf{P}_i^2}{2m}+\sum_{\{i,j\}}\phi(\mathbf{r}_{ij})-\sum_{\{i,j\}}J(\mathbf{r}_{ij})\mathbf{S}_i\cdot\mathbf{S}_j -D\sum_{i=1}^{N}(S_i^z)^2-g\mu_B H\sum_{i=1}^{N}S_i^z+\sum_{\{i,j\}}\boldsymbol{\lambda}\cdot\mathbf{r}_{ij}\ , \tag{1}$$

where the first term represents the kinetic energy of the N ions of mass m and the second term is the elastic potential energy between first neighbors. The third term represents the antiferromagnetic exchange between nearest neighbors ($J<0$) which depends on the relative distance between neighboring ions situated at the pair of sites i and j. The parameter D represents the single-ion uniaxial anisotropy and H is the static magnetic field applied along the easy axis. The last term of Eq. (1) represents the work done by the external force $\boldsymbol{\lambda}$, which is applied along every line of ions of the crystal.

For simplicity we assume a harmonic potential given by

$$\phi(\mathbf{r}_{ij})=\phi_0+\frac{\alpha}{2}(\,|\,\mathbf{r}_{ij}\,|\,-a_0)^2\ , \tag{2}$$

where ϕ_0 and α are positive constants and a_0 is the average distance between neighbor spins at a given temperature T_0. Expanding this potential around the equilibrium positions of the ions at a temperature T, we obtain

$$\phi=\phi_0+\frac{\alpha}{2}\left\{(a-a_0)^2+2(a-a_0)\mu_{ij}^{\alpha}+(\mu_{ij}^{\alpha})^2 +\left[1-\left[\frac{a_0}{a}\right]\right]\sum_{\beta(\neq\alpha)}(\mu_{ij}^{\beta})^2\right\}+\cdots\ , \tag{3}$$

where μ_{ij}^{α} ($\alpha = x,y,z$) represents the Cartesian component of the relative deviation from equilibrium positions of the ions and a is the average distance between neighboring spins at temperature T. Neglecting the last term in (3), which means disregarding the shear forces, we arrive at the Baker-Essam model. In this case both the elastic and magnetic couplings depend only on the longitudinal component of the relative positions of the ions. For the exchange interaction we also assume that

$$J(\mathbf{r}_{ij}) = J_0 + J_1(|\xi_{ij}| - a_0) , \tag{4}$$

where $J_0 < 0$ and $J_1 > 0$. According to the Baker-Essam model

$$\xi_{ij} = \mathbf{r}_{ij} \cdot \mathbf{l}_{ij} , \tag{5}$$

where $\mathbf{l}_{ij}$ is a unit vector in the direction of the corresponding ions located at the sites i and j in the rigid lattice. Applying the following unitary transformation[8]

$$U = \prod_l \exp\left[i \frac{J_1}{\alpha} P_l \sum_{m(<l)} \mathbf{S}_m \cdot \mathbf{S}_{m+1} \right] , \tag{6}$$

where $[x_j, P_l] = i\delta_{jl}$, to the Hamiltonian given by Eq. (1), we decouple the Hamiltonian into parts which depend separately on the spin and lattice degrees of freedom. This is possible because within the Baker-Essam model, every line of spins is elastically independent of the others[9] and we apply the above transformation to every single line. In this way we obtain the following effective spin Hamiltonian:

$$\mathcal{H}^{\text{eff}} = -\sum_{\{i,j\}} [J(\lambda)\mathbf{S}_i \cdot \mathbf{S}_j + A(\mathbf{S}_i \cdot \mathbf{S}_j)^2] - D \sum_{i=1}^{N} (S_i^z)^2 - g\mu_B H \sum_{i=1}^{N} S_i^z , \tag{7}$$

where

$$J(\lambda) = J_0 - \frac{J_1 \lambda}{\alpha} \text{ and } A = \frac{J_1^2}{2\alpha} > 0 . \tag{8}$$

We observe the presence of the biquadratic coupling that appears naturally as a consequence of the spin-lattice interaction, and the explicit dependence of the exchange parameter on the external force.

The phase diagram for this compressible antiferromagnetic model can be obtained from the magnetic free energy $G(T,H,\lambda,N)$. We use the Bogoliubov's inequality

$$G(\mathcal{H}^{\text{eff}}) \le G_0 + \langle \mathcal{H}^{\text{eff}} - \mathcal{H}_0 \rangle_0 = \overline{G} \tag{9}$$

to obtain an upper bound $\overline{G}$ for the true free energy of the system. Taking the following trial Hamiltonian:

$$\mathcal{H}_0 = -\sum_{\alpha} \left[\sum_{i=1}^{N/2} K_A^{\alpha} S_{iA}^{\alpha} + \sum_{j=1}^{N/2} K_B^{\alpha} S_{jB}^{\alpha} \right] , \tag{10}$$

where $K_{A,B}^{\alpha}$ ($\alpha = x,y,z$) are the six variational parameters for both sublattices A and B, we obtain an upper bound for G when we minimize $\overline{G}$ with respect to the parameters $K_{A,B}^{\alpha}$. It is straightforward to obtain the following expressions for G_0 and for the mean values of interest:

$$G_0 = -\frac{N}{2\beta} \{ \ln[1 + 2\cosh(\beta\lambda_A)] + \ln[1 + 2\cosh(\beta\lambda_B)] \} , \tag{11}$$

where

$$\beta = (k_B T)^{-1} \text{ and } \lambda_{A,B} = \left[\sum_{\alpha} (K_{A,B}^{\alpha})^2 \right]^{1/2} , \tag{12}$$

$$\langle S_{A,B}^{\alpha} \rangle = \frac{2\sinh(\beta\lambda_{A,B})}{1 + 2\cosh(\beta\lambda_{A,B})} \frac{K_{A,B}^{\alpha}}{\lambda_{A,B}} , \tag{13}$$

$$\langle (S_{A,B}^{\alpha})^2 \rangle = \frac{2}{\beta[1 + 2\cosh(\beta\lambda_{A,B})]} \left\{ \beta \left[\frac{K_{A,B}^{\alpha}}{\lambda_{A,B}} \right]^2 \cosh(\beta\lambda_{A,B}) + \frac{1}{\lambda_{A,B}} \left[1 - \left[\frac{K_{A,B}^{\alpha}}{\lambda_{A,B}} \right]^2 \right] \sinh(\beta\lambda_{A,B}) \right\} \tag{14}$$

$$\langle S_{A,B}^{\alpha} S_{A,B}^{\gamma} \rangle = \frac{2}{\beta[1 + 2\cosh(\beta\lambda_{A,B})]} \frac{K_{A,B}^{\alpha} K_{A,B}^{\gamma}}{\lambda_{A,B}^2} \left[\beta\cosh(\beta\lambda_{A,B}) - \frac{\sinh(\beta\lambda_{A,B})}{\lambda_{A,B}} \right], \quad \alpha \ne \gamma . \tag{15}$$

With these expressions we can write an expression for $\overline{G}$ Eq. (9), as a function of the parameters $K_{A,B}^{\alpha}$. Minimizing $\overline{G}$ on these parameters we obtain a set of six equations that can be solved numerically to determine the complete phase diagram.

III. PHASE DIAGRAM

Let us initially consider the phase transitions that occur at $T = 0$ K. The energies per spin in the antiferromagnetic phase (AF), spin-flop phase (SF), and paramagnetic phase (P) are given, respectively, by

$$U_{\text{AF}} = 3J - 3A - D , \tag{16}$$

$$U_{\text{SF}} = -3J\cos(2\theta) - 3A\cos^2(2\theta) - g\mu_B H\cos\theta - D\cos^2\theta , \tag{17}$$

$$U_{\text{P}} = -3J - 3A - g\mu_B H - D , \tag{18}$$

where we have considered a simple-cubic lattice with $S = 1$ and J is given by Eq. (8). In Eq. (17), θ is the angle that both sublattices form with the external field in the spin-flop phase.

The phase transitions are determined when the free energies of different phases become equal for a given set of parameters. The transition between antiferromagnetic and paramagnetic phases is of first order and the critical field is given by

$$g\mu_B H_{\text{AF-P}} = -6J \ . \tag{19}$$

This critical field is independent of A and D. In order to determine the other transitions we have to minimize U_{SF} with respect to θ:

$$\frac{\partial U_{\text{SF}}}{\partial \theta} = 48A\cos^3\theta + (2D + 12J - 24A)\cos\theta + g\mu_B H \ . \tag{20}$$

The transition between antiferromagnetic and spin-flop phases are obtained considering the simultaneous solutions of the following equations:

$$U_{\text{AF}} = U_{\text{SF}} \text{ and } \frac{\partial U_{\text{SF}}}{\partial \theta} = 0 \ . \tag{21}$$

This system has only one solution that has a physical meaning and this transition is always of first order. On the other hand the transition between the spin-flop phase and the paramagnetic phase, given by the solutions of the following equations:

$$U_{\text{SF}} = U_{\text{P}} \text{ and } \frac{\partial U_{\text{SF}}}{\partial \theta} = 0 \ , \tag{22}$$

can be of first or second order depending on the values of the parameters A and D. The critical field in the case of second-order transition can be obtained analytically and is given by

$$g\mu_B H_{\text{SF-P}} = -12J - 24A - 2D \ . \tag{23}$$

The results for the first-order transitions can be only obtained numerically and the values for the critical field are bigger than those given by Eq. (23).

In Fig. 1 we present a diagram in the space A, D, H. In region I the SF-P second-order phase transitions occur. In region II we have the SF-P first-order phase transitions. In region III we do not have the spin-flop phase. This region is characteristic of a metamagnetic model and the antiferromagnetic to paramagnetic phase transition is always of first order. This critical field is independent of A and D [see Eq. (19)]. In Fig. 2 we exhibit a diagram of the critical fields as a function of the parameter A for fixed values of J and D. Of course, this figure is a projection in the H-A plane of Fig. 1. We sketch it separately because in Fig. 1 we do not present the AF-SF phase transition as a matter of clearness. We observe that for $A > A_c$ the system does not present the spin-flop phase, that is, it behaves like a metamagnetic model.

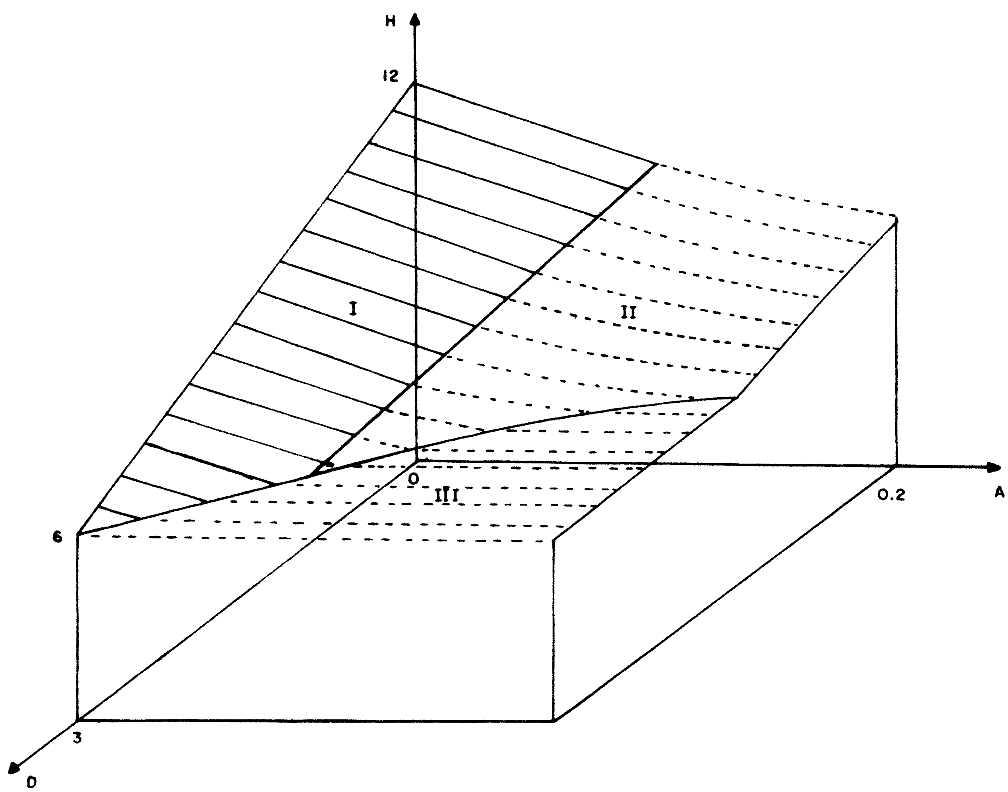

FIG. 1. H-A-D diagram at $T=0$. The solid lines in region I represent the SF-P second-order phase transitions. The dashed lines in region II represent the transitions between SF and P phases which are of first order while in region III they represent the first-order AF-P phase transitions. We have $J=-1$ and the parameters A and D are dimensionless.

The result of the minimization of the variational free energy described in the Sec. II produces three different phases, namely

$$\text{antiferromagnetic (AF):}\begin{cases}\langle S_A^x\rangle=\langle S_B^x\rangle=0\ ,\\ \langle S_A^z\rangle\neq\langle S_B^z\rangle\neq 0\ ,\end{cases}\tag{24}$$

$$\text{spin-flop (SF):}\begin{cases}\langle S_A^x\rangle=-\langle S_B^x\rangle>0\ ,\\ \langle S_A^z\rangle=\langle S_B^z\rangle>0\ ,\end{cases}\tag{25}$$

$$\text{paramagnetic (P):}\begin{cases}\langle S_A^x\rangle=\langle S_B^x\rangle=0\ ,\\ \langle S_A^z\rangle=\langle S_B^z\rangle\geq 0\ ,\end{cases}\tag{26}$$

where we have defined the y axis such that $\langle S_A^y\rangle=\langle S_B^y\rangle=0$. The effective spin Hamiltonian given by Eq. (7) is invariant with respect to rotations in the x-y plane.

In Fig. 3 we present the phase diagram of a compressible antiferromagnetic model for some chosen values of external force. As it is expected, the critical fields at $T=0$ and the Néel temperature increase linearly with the forces due to the harmonic approximation considered for the elastic potential energy in our model. For instance, the Néel temperature (T_N) can be analytically determined and it is given by

$$T_N=\frac{-12J-4A+2D}{3}\ .\tag{27}$$

This behavior seems to be verified in some compressible antiferromagnets.[2] In Fig. 3 we also exhibit the evolution of the bicritical point as a function of external force. The temperature and magnetic field associated with the bicritical point increase with the external tensions. We have also considered the evolution of bicritical point as a function of the biquadratic parameter for fixed values of the single-ion uniaxial anisotropy. We have seen that as A increases the region in the plane H versus T associated with the spin-flop phase diminishes. In this way the biquadratic exchange parameter A is similar to the single-ion parameter D and the global phase diagram looks like the uniaxial antiferromagnets.

IV. LOW-TEMPERATURE SPIN-FLOP–PARAMAGNETIC TRANSITION

In this section we determine the spin-flop–paramagnetic transition at very low temperatures. From the experimental point of view this is an interesting problem because we can study spin waves as a function of the pressure and of the biquadratic exchange parameter in the low-temperature region.

We consider the compressible antiferromagnetic model in its paramagnetic phase as being described by the effective spin Hamiltonian given by Eq. (7) of Sec. II. We define raising and lowering spin operators of the form $S_l^{\pm}=S_l^x\pm iS_l^y$ for every lattice point. Now we introduce the Holstein-Primakoff representation to write the operators $S_l^{\pm}$ and S_l^z in terms of spin deviations operators at each lattice site. The next step is to switch to the Fourier representation and expand the Hamiltonian up to terms of order S^{-1}. If we neglect the contribution of terms with more than four magnon operators, it is very easy to use Wick's theorem to linearize the equation of motion in the Heisenberg representation for the spin-wave operator $a_{\mathbf{k}}$, where $\mathbf{k}$ are vectors of the first Brillouin zone. In this way we obtain the following effective-energy spectrum $\varepsilon_{\mathbf{k}}(T,H)$ of the magnons:

$$\varepsilon_{\mathbf{k}}(T,H)=A_{\mathbf{k}}+\frac{1}{NS}\sum_{\mathbf{q}}F_{\mathbf{kq}}\langle a_{\mathbf{q}}^{\dagger}a_{\mathbf{q}}\rangle\ ,\tag{28}$$

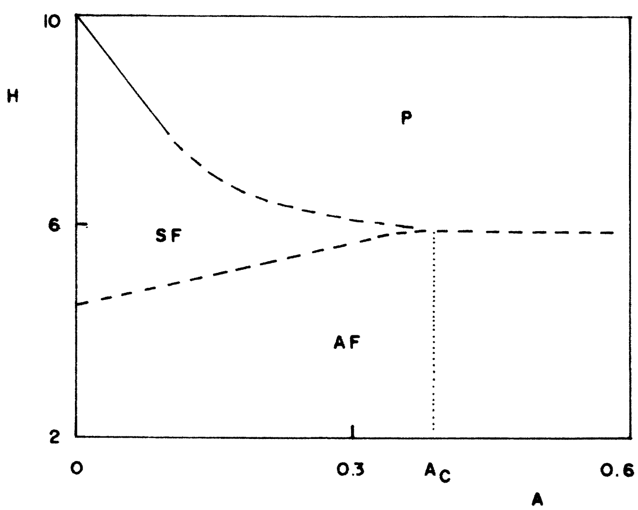

FIG. 2. Critical field as a function of the biquadratic exchange parameter A. The solid line is a line of second-order transition and the dashed lines represent the first-order transitions. We see the three phases, antiferromagnetic (AF), spin-flop (SF), and paramagnetic (P), of an antiferromagnet. We have $T=0$ K, $J=-1$, $D=1$, and $A_c=0.378$. All parameters are dimensionless.

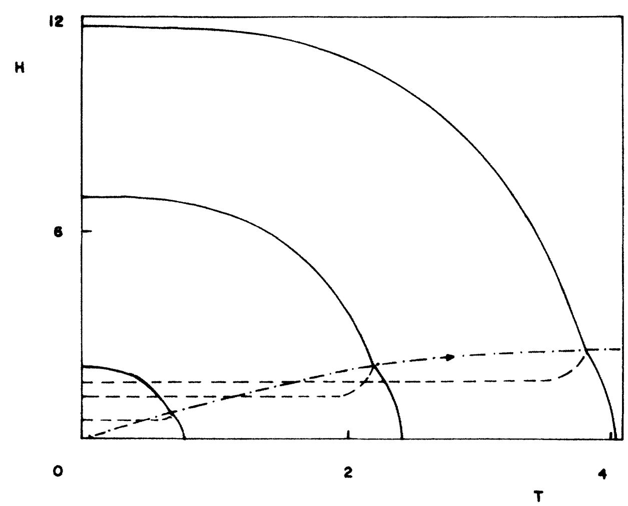

FIG. 3. Phase diagram of a compressible antiferromagnetic model for some values of external force. The dot-dashed line indicates the evolution of bicritical point with external force. We have $D=0.1$, $A=0.01$, and $J(\lambda)=-0.2$, -0.6, and -1.0. All parameters are dimensionless.

where

$$A_{\mathbf{k}} = [-ZSJ(\lambda) + 2ZA(1-S)S^2](\gamma_{\mathbf{k}} - 1) + (2S-1)D + g\mu_B H , \quad (29)$$

and

$$F_{\mathbf{kq}} = -ZJS[1 + \gamma_{\mathbf{k}-\mathbf{q}} - \gamma_{\mathbf{k}} - \gamma_{\mathbf{q}}] - 2AS^3 Z[S(1-\gamma_{\mathbf{k}} - \gamma_{\mathbf{q}}) + 2\gamma_{\mathbf{k}+\mathbf{q}} + 3\gamma_{\mathbf{k}-\mathbf{q}}] - 4DS , \quad (30)$$

where $\gamma_{\mathbf{k}} = (1/Z)\sum_{\delta} e^{-i\mathbf{k}\cdot\delta}$ is the structure factor for the Z nearest neighbors of a given ion. We also have that

$$\langle a_{\mathbf{k}}^{\dagger} a_{\mathbf{k}} \rangle = (e^{\beta\varepsilon_{\mathbf{k}}} - 1)^{-1} . \quad (31)$$

is the boson occupation number.

The spin-flop–paramagnetic critical field $H_c(T)$ is determined by the limit of stability of the paramagnetic phase, namely by the equation $\varepsilon_{\mathbf{k}_0}(T, H_c) = 0$, where the vector $\mathbf{k}_0$ labels the corners of the first Brillouin zone. The asymptotic form of the critical field at low temperatures is given by

$$g\mu_B H_c(T) = A_0 - A_{3/2} T^{3/2} - A_{5/2} T^{5/2} \ldots , \quad (32)$$

where

$$A_0 = -2ZSJ(\lambda) + 4ZS^2(1-S)A - D(2S-1) , \quad (33)$$

$$A_{3/2} = -\frac{1}{\pi^2}\{Z[J(\lambda) + 10S^2 A] + D\} \times \Gamma\left[\frac{3}{2}\right] \xi\left[\frac{3}{2}\right] \left[\frac{k_B}{a}\right]^{3/2} , \quad (34)$$

$$A_{5/2} = -\frac{1}{\pi^2}\frac{1}{4}\left[\frac{ZJ(\lambda)}{6} + \frac{D}{2} + \frac{5ZAS^2}{3}\right] \times \Gamma\left[\frac{5}{2}\right] \xi\left[\frac{5}{2}\right] \left[\frac{k_B}{a}\right]^{5/2} . \quad (35)$$

In these expressions, $\Gamma(n)$ is the γ function,

$$\xi(\alpha) = \sum_{n=1}^{\infty} (n)^{-\alpha} ,$$

$$a = \frac{ZS}{6}[-J(\lambda) + 2S(1-S)A] , \quad (36)$$

and k_B is the Boltzmann constant.

We would like to stress that our spin-wave expression for the critical field at $T=0$ reduces to the mean field one for high-spin values, i.e.,

$$g\mu_B H_c(0) = -2ZSJ(\lambda) - 4ZAS^3 - 2DS . \quad (37)$$

If we consider this last equation for a simple-cubic lattice with $S=1$, we recover our Eq. (23), obtained in the last section.

Therefore, the external pressures and biquadratic exchange term do not change the asymptotic $T^{3/2}$ Bloch law for the canted-paramagnetic phase boundary at very low temperatures. A detailed analysis of the experimental results[1,2] at very low temperatures is necessary in order to determine the validity of our result.

V. CONCLUSIONS

We have considered in this work a compressible antiferromagnetic model with an applied field directed along the axis of anisotropy. Our magnetic elastic model is due to Baker and Essam where the shear forces are disregarded. We obtain an effective spin Hamiltonian, where the exchange terms are dependent on external forces and we now have a biquadratic exchange coupling between spins. Through Bogoliubov's inequality we have determined a variational free energy for the system of spins. A global phase diagram in the plane H (magnetic field) versus T (temperature) was determined for different values of external forces. A novel feature was observed in the spin-flop–paramagnetic phase boundary: it can be of first or second order depending on the values of single-ion uniaxial anisotropy and biquadratic exchange parameter. We also have shown that there is a critical value for the biquadratic exchange parameter where the system becomes metamagnetic. The critical fields obtained at $T=0$ K and the Néel temperature increase linearly with the pressure as it is expected to experimentally. The behavior of the bicritical point as a function of the biquadratic term A is similar to the one observed as a function of the single-ion uniaxial term D. As A or D increases, the region of the phase diagram occupied by the spin-flop phase diminishes. Finally, we have performed spin-wave calculations on the effective spin Hamiltonian with exchange terms depending on external forces and with biquadratic coupling. We have shown that at very low-temperatures the canted-paramagnetic phase boundary follows a $T^{3/2}$ Bloch law.

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