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Ferrimagnetism of MnV_2O_4 spinel

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Abstract

The spinel MnV_2O_4 is a two-sublattice ferrimagnet, with site A occupied by the Mn^{2+} ion and site B by the V^{3+} ion. The magnon of the system, the transversal fluctuation of the total magnetization, is a complicated mixture of the sublattice A and B transversal magnetic fluctuations. As a result, the magnons' fluctuations suppress in a different way the manganese and vanadium sublattice magnetization and one obtains two phases. At low temperature ($0, T^*$) the sublattice Mn magnetization and sublattice V magnetization contribute to the magnetization of the system, while at a high temperature (T^*, T_N), the vanadium sublattice magnetization is suppressed by magnon fluctuations, and only the manganese ions have non-zero spontaneous magnetization. A modified spin-wave theory is developed to describe the two phases and to calculate the magnetization as a function of temperature. The anomalous $M(T)$ curve reproduces the experimentally obtained zero-field-cooled (ZFC) magnetization.

This paper is inspired from experimental measurements of the zero-field-cooled (ZFC) magnetization of MnV_2O_4 [1–3]. The profile of the experimental curve reproduces the anomalous magnetization curve predicted by Néel [6, 7] for ferrimagnets with equal sublattice spins. This stimulates us to model the manganese vanadate spinel in the spirit of Néel's theory. By comparing and contrasting ZFC and field-cooled (FC) magnetization one gains an insight into the magnetism of the manganese vanadate oxide.

The spinel MnV_2O_4 is a two-sublattice ferrimagnet, with site A occupied by the Mn^{2+} ion, which is in the $3d^5$ high-spin configuration with quenched orbital angular momentum, which can be regarded as a simple $s = 5/2$ spin. The B site is occupied by the V^{3+} ion, which takes the $3d^2$ high-spin configuration in the triply degenerate t_{2g} orbital and has orbital degrees of freedom. The measurements show that the setting in of the magnetic order is at the Néel temperature $T_N = 56.5$ K [1], and that the magnetization has a maximum near $T^* = 53.5$ K. Below this temperature the magnetization sharply decreases and goes to zero when the temperature approaches zero. A second transition is observed at T^* from collinear ferrimagnetism to triangular, accompanied by a cubic-to-tetragonal distortion [1–5]. There is a thermodynamic signature that this is a first-order transition [5].

We consider a system which obtains its magnetic properties from Mn and V magnetic moments. It is shown that the true magnons in this system, which are the transversal fluctuations corresponding to the total magnetization, are complicated mixtures of the Mn and V transversal fluctuations. The magnons interact with manganese and vanadium ions in a

different way, and the magnon fluctuations suppress the Mn and V sublattice magnetization at different temperatures. As a result, the ferrimagnetic phase is divided into two phases: in the low temperature phase $0 < T < T^*$ the sublattice Mn magnetization and sublattice V magnetization contribute to the magnetization of the system, while in the high temperature phase (T^*, T_N), the vanadium sublattice magnetization is suppressed by magnon fluctuations, and only the manganese ions have non-zero spontaneous magnetization. A modified spin-wave theory is developed to describe the two phases and to calculate the magnetization as a function of temperature. The anomalous $M(T)$ curve reproduces the experimentally obtained ZFC magnetization [2, 3].

Because of the strong spin–orbital interaction it is convenient to consider jj coupling with $\mathbf{J}^A = \mathbf{S}^A$ and $\mathbf{J}^B = \mathbf{L}^B + \mathbf{S}^B$. The sublattice A total angular momentum is $j_A = s_A = 5/2$, while the sublattice B total angular momentum is $j_B = l_B + s_B$, with $l_B = 3$ and $s_B = 1$ [1]. Then the g factor for the sublattice A is $g_A = 2$ and the atomic value of g_B is $g_B = \frac{5}{4}$. The sublattice A magnetic order is antiparallel to the sublattice B one, and the saturated magnetization is $\sigma = 2\frac{5}{2} - \frac{5}{4}4 = 0$ in agreement with the experimental finding that the magnetization goes to zero when the temperature approaches zero.

The Hamiltonian of the system is

$$H = -\kappa_A \sum_{\langle\langle ij \rangle\rangle_A} \mathbf{J}_i^A \cdot \mathbf{J}_j^A - \kappa_B \sum_{\langle\langle ij \rangle\rangle_B} \mathbf{J}_i^B \cdot \mathbf{J}_j^B + \kappa \sum_{\langle ij \rangle} \mathbf{J}_i^A \cdot \mathbf{J}_j^B, \quad (1)$$

where the sums are over all sites of a three-dimensional cubic lattice: $\langle i, j \rangle$ denotes the sum over the nearest neighbors, while $\langle\langle i, j \rangle\rangle_{A(B)}$ denotes the sum over the sites of the A(B) sublattice. The first two terms describe the ferromagnetic Heisenberg intra-sublattice exchange $\kappa_A > 0$, $\kappa_B > 0$, while the third term describes the inter-sublattice exchange which is antiferromagnetic $\kappa > 0$.

To proceed we use the Holstein–Primakoff representation of the total angular momentum vectors $\mathbf{J}_j^A(a_j^+, a_j)$ and $\mathbf{J}_j^B(b_j^+, b_j)$, where a_j^+, a_j and b_j^+, b_j are Bose fields. In terms of these fields and keeping only the quadratic and quartic terms, the effective Hamiltonian equation (1) adopts the form, $H = H_2 + H_4$, where

$$H_2 = j_A \kappa_A \sum_{\langle\langle ij \rangle\rangle_A} \left(a_i^+ a_i + a_j^+ a_j - a_j^+ a_i - a_i^+ a_j \right) + j_B \kappa_B \sum_{\langle\langle ij \rangle\rangle_B} \left(b_i^+ b_i + b_j^+ b_j - b_j^+ b_i - b_i^+ b_j \right) + \kappa \sum_{(ij)} \left[j_A b_j^+ b_j + j_B a_i^+ a_i - \sqrt{j_A j_B} \left(a_i^+ b_j^+ + a_i b_j \right) \right] \quad (2)$$

$$H_4 = \frac{1}{4} \kappa_A \sum_{\langle\langle ij \rangle\rangle_A} \left[a_i^+ a_j^+ (a_i - a_j)^2 + (a_i^+ - a_j^+)^2 a_i a_j \right] + \frac{1}{4} \kappa_B \sum_{\langle\langle ij \rangle\rangle_B} \left[b_i^+ b_j^+ (b_i - b_j)^2 + (b_i^+ - b_j^+)^2 b_i b_j \right] + \frac{1}{4} \kappa \sum_{(ij)} \left[\sqrt{\frac{j_A}{j_B}} \left(a_i b_j^+ b_j b_j + a_i^+ b_j^+ b_j^+ b_j \right) + \sqrt{\frac{j_B}{j_A}} \left(a_i^+ a_i a_i b_j + a_i^+ a_i^+ a_i b_j^+ \right) - 4 a_i^+ a_i b_j^+ b_j \right] \quad (3)$$

and the terms without operators are dropped.

The next step is to represent the Hamiltonian in the Hartree–Fock approximation $H \approx H_{\text{HF}} = H_{\text{cl}} + H_q$, where

$$H_{\text{cl}} = 12N\kappa_A j_A^2 (u_A - 1)^2 + 12N\kappa_B j_B^2 (u_B - 1)^2 + 6N\kappa j_A j_B (u - 1)^2 \quad (4)$$

and $N = N_A = N_B$ is the number of sites on a sublattice. The Hamiltonian H_q can be obtained from the Hamiltonian equation (2) replacing κ_A with $\kappa_A u_A$, κ_B with $\kappa_B u_B$ and κ with κu , where u_A , u_B and u are Hartree–Fock parameters, to be determined self-consistently. It is convenient to rewrite the Hamiltonian in momentum space representation:

$$H_q = \sum_{k \in B_r} \left[\varepsilon_k^a a_k^+ a_k + \varepsilon_k^b b_k^+ b_k - \gamma_k \left(a_k^+ b_k^+ + b_k a_k \right) \right], \quad (5)$$

where the wavevector k runs over the reduced first Brillouin zone B_r of a cubic lattice. The dispersions are given by the equalities

$$\begin{aligned} \varepsilon_k^a &= 4j_A \kappa_A u_A \varepsilon_k + 6j_B \kappa u \\ \varepsilon_k^b &= 4j_B \kappa_B u_B \varepsilon_k + 6j_A \kappa u \\ \gamma_k &= 2\kappa u \sqrt{j_A j_B} \left(\cos k_x + \cos k_y + \cos k_z \right) \end{aligned} \quad (6)$$

with $\varepsilon_k = 6 - \cos(k_x + k_y) - \cos(k_x - k_y) - \cos(k_x + k_z) - \cos(k_x - k_z) - \cos(k_y + k_z) - \cos(k_y - k_z)$.

To diagonalize the Hamiltonian one introduces new Bose fields $\alpha_k, \alpha_k^+, \beta_k, \beta_k^+$ by means of the transformation

$$a_k = u_k \alpha_k + v_k \beta_k^+, \quad b_k = u_k \beta_k + v_k \alpha_k^+ \quad (7)$$

with coefficients u_k and v_k real functions of the wavevector k :

$$u_k = \sqrt{\frac{1}{2} \left(\frac{\varepsilon_k^a + \varepsilon_k^b}{\sqrt{(\varepsilon_k^a + \varepsilon_k^b)^2 - 4\gamma_k^2}} + 1 \right)}, \quad (8)$$

$v_k = \text{sign}(\gamma_k) \sqrt{u_k^2 - 1}$. The transformed Hamiltonian adopts the form

$$H_q = \sum_{k \in B_r} \left(E_k^\alpha \alpha_k^+ \alpha_k + E_k^\beta \beta_k^+ \beta_k + E_k^0 \right), \quad (9)$$

with new dispersions $E_k^\alpha = E_k^+$, $E_k^\beta = E_k^-$, where

$$E_k^\pm = \frac{1}{2} \left[\sqrt{(\varepsilon_k^a + \varepsilon_k^b)^2 - 4\gamma_k^2} \pm (\varepsilon_k^a - \varepsilon_k^b) \right] \quad (10)$$

and vacuum energy

$$E_k^0 = \frac{1}{2} \left[\sqrt{(\varepsilon_k^a + \varepsilon_k^b)^2 - 4\gamma_k^2} - \varepsilon_k^b - \varepsilon_k^a \right]. \quad (11)$$

To obtain the system of equations for the Hartree–Fock parameters we consider the free energy of a system with Hamiltonian H_{HF} equations (4) and (9):

$$\begin{aligned} \mathcal{F} &= 12\kappa_A j_A^2 (u_A - 1)^2 + 12\kappa_B j_B^2 (u_B - 1)^2 \\ &+ 6\kappa j_A j_B (u - 1)^2 + \frac{1}{N} \sum_{k \in B_r} E_k^0 \\ &+ \frac{1}{\beta N} \sum_{k \in B_r} \left[\ln(1 - e^{-\beta E_k^\alpha}) + \ln(1 - e^{-\beta E_k^\beta}) \right]. \end{aligned} \quad (12)$$

Then the three equations $\partial \mathcal{F} / \partial u_A = 0$, $\partial \mathcal{F} / \partial u_B = 0$ and $\partial \mathcal{F} / \partial u = 0$ adopt the form

$$\begin{aligned} u_A &= 1 - \frac{1}{6j_A} \frac{1}{N} \sum_{k \in B_r} \varepsilon_k \left[u_k^2 n_k^\alpha + v_k^2 n_k^\beta + v_k^2 \right] \\ u_B &= 1 - \frac{1}{6j_B} \frac{1}{N} \sum_{k \in B_r} \varepsilon_k \left[v_k^2 n_k^\alpha + u_k^2 n_k^\beta + v_k^2 \right] \\ u &= 1 - \frac{1}{N} \sum_{k \in B_r} \left[\frac{1}{2j_A} \left(u_k^2 n_k^\alpha + v_k^2 n_k^\beta + v_k^2 \right) \right. \\ &+ \frac{1}{2j_B} \left(v_k^2 n_k^\alpha + u_k^2 n_k^\beta + v_k^2 \right) \\ &\left. - \frac{2}{3} \kappa u \left(1 + n_k^\alpha + n_k^\beta \right) \frac{(\cos k_x + \cos k_y + \cos k_z)^2}{\sqrt{(\varepsilon_k^a + \varepsilon_k^b)^2 - 4\gamma_k^2}} \right], \end{aligned} \quad (13)$$

where n_k^α and n_k^β are the Bose functions of α and β excitations. Hartree–Fock parameters, the solution of the system of equations (13), are positive functions of T/κ , $u_A(T/\kappa) > 0$,

$u_B(T/\kappa) > 0$ and $u(T/\kappa) > 0$. Utilizing these functions, one can calculate the spontaneous magnetization $M^A = \langle J_3^A \rangle$ and $M^B = \langle J_3^B \rangle$ of Mn and V ions, where J_3^A and J_3^B are the third components of the \mathbf{J}^A and \mathbf{J}^B vectors, respectively. In terms of the Bose functions of the α and β excitations they adopt the form

$$\begin{aligned} M^A &= j_A - \frac{1}{N} \sum_{k \in B_r} \left[u_k^2 n_k^\alpha + v_k^2 n_k^\beta + v_k^2 \right] \\ M^B &= -j_B + \frac{1}{N} \sum_{k \in B_r} \left[v_k^2 n_k^\alpha + u_k^2 n_k^\beta + v_k^2 \right]. \end{aligned} \quad (14)$$

The magnon excitations in the effective theory are a complicated mixture of the sublattices' A and B transversal fluctuations. As a result, the magnon fluctuations suppress in a different way the sublattice Mn magnetization and sublattice V magnetization. Quantitatively, this depends on the coefficients u_k and v_k in equation (14). At characteristic temperature T^* V spontaneous magnetization becomes equal to zero, while Mn spontaneous magnetization is still non-zero. Above T^* the system of equations (13) has no solution and one has to modify the spin-wave theory.

Once suppressed, the sublattice V magnetization cannot be restored increasing the temperature above T^* . To formulate this mathematically we modify the spin-wave theory using the idea of the description of the paramagnetic phase of 2D ferromagnets ($T > 0$) by means of modified spin-wave theory [8, 9]. We consider a two-sublattice system and to enforce the magnetization on the two sublattices to be equal to zero in paramagnetic phase we introduce two parameters λ_A and λ_B [10]. The new Hamiltonian is obtained from the old one (equation (1)) adding two new terms:

$$\hat{H} = H - \sum_{i \in A} \lambda_A J_{3i}^A + \sum_{i \in B} \lambda_B J_{3i}^B. \quad (15)$$

In momentum space, the Hamiltonian adopts the form equation (5) with new dispersions $\hat{\varepsilon}_k^a = \varepsilon_k^a + \lambda_A$ and $\hat{\varepsilon}_k^b = \varepsilon_k^b + \lambda_B$, where the old dispersions are given by equalities (6). We utilize the same transformation equation (7) with coefficients \hat{u}_k and \hat{v}_k which depend on the new dispersions in the same way as the old ones depend on the old dispersions equation (8). In terms of the α_k and β_k bosons, the Hamiltonian \hat{H}_q adopts the form equation (9) with dispersions \hat{E}_k^α and \hat{E}_k^β , which can be written in the form equation (10), replacing ε_k^a and ε_k^b with $\hat{\varepsilon}_k^a$ and $\hat{\varepsilon}_k^b$.

We have to do some assumptions for parameters λ_A and λ_B to ensure the correct definition of the two-boson theory. For that purpose, it is convenient to represent the parameters in the form $\lambda_A = 6\kappa u j_B (\mu_A - 1)$ $\lambda_B = 6\kappa u j_A (\mu_B - 1)$. In terms of the parameters μ_A and μ_B , the dispersions are $\hat{\varepsilon}_k^a = 4j_A \kappa_A u_A \varepsilon_k + 6\kappa u j_B \mu_A$, $\hat{\varepsilon}_k^b = 4j_B \kappa_B u_B \varepsilon_k + 6\kappa u j_A \mu_B$. The conventional spin-wave theory is reproduced when $\mu_A = \mu_B = 1$ ($\lambda_A = \lambda_B = 0$). We assume μ_A and μ_B to be positive ($\mu_A > 0, \mu_B > 0$). Then, $\hat{\varepsilon}_k^a > 0, \hat{\varepsilon}_k^b > 0$, for all values of the wavevector k if the Hartree–Fock parameters are positive too. The Bose theory is well defined if $E_k^\alpha \geq 0, E_k^\beta \geq 0$. This comes true if $\mu_A \mu_B \geq 1$. In the case $\mu_A \mu_B > 1$, both

α_k and β_k bosons are gapped excitations. In the particular case $\mu_A \mu_B = 1$, long-range excitations (magnons) emerge in the system.

We introduced the parameters λ_A and λ_B (μ_A, μ_B) to enforce the sublattice A and B spontaneous magnetizations to be equal to zero in the paramagnetic phase. We find out the parameters μ_A and μ_B , as well as the Hartree–Fock parameters, as functions of temperature, solving the system of five equations, equations (13) and the equations $M^A = M^B = 0$, where the ordered moments have the same representation as equation (14) but with coefficients \hat{u}_k, \hat{v}_k , and dispersions $\hat{E}_k^\alpha, \hat{E}_k^\beta$ in the expressions for the Bose functions. The numerical calculations show that, for high enough temperature, $\mu_A \mu_B > 1$. When the temperature decreases the product $\mu_A \mu_B$ decreases, remaining larger than 1. The temperature at which the product becomes equal to 1 ($\mu_A \mu_B = 1$) is the Néel temperature. Below T_N , the spectrum contains long-range (magnon) excitations, thereupon $\mu_A \mu_B = 1$. It is convenient to represent the parameters in the following way:

$$\mu_A = \mu, \quad \mu_B = 1/\mu. \quad (16)$$

In the ordered phase magnon excitations are the origin of the suppression of the magnetization. Near zero temperature their contribution is small and at zero temperature Mn and V spontaneous magnetization reach saturation. Increasing the temperature magnon fluctuations suppress the sublattice Mn magnetization and sublattice V magnetization in different ways. At T^* the V spontaneous magnetization becomes equal to zero. Increasing the temperature above T^* , the vanadium sublattice magnetization should be zero. This is why we impose the condition $M^B(T) = 0$ if $T > T^*$. For temperatures above T^* , the parameter μ and the Hartree–Fock parameters are solutions of a system of four equations, equations (13) and the equation $M^B = 0$. We utilize the obtained function $\mu(T), u_A(T), u_B(T), u(T)$ to calculate the spontaneous magnetization M^A of the Mn ions as a function of the temperature. Above T^* , $M^A(T)$ is equal to the magnetization of the system. It is important to stress that the system of equations for λ_A, λ_B and Hartree–Fock parameters is equivalent to the system of equations which one obtains treating the paramagnetic phase of a system with Hamiltonian equation (1) by means of the Schwinger-boson mean field theory. The advantage of the modified spin-wave theory is the possibility of extending the calculations for temperatures below the Néel temperature. Introducing two λ parameters one captures the important physical feature of the system, namely the magnons are long-range excitations below the critical temperature, and they open a gap above T_N .

We consider a two-sublattice ferrimagnet with Mn ions on sublattice A and V ions on sublattice B. The sublattice A total angular moment is $j_A = s_A = 5/2$ and g factor $g_A = 2$. The sublattice B total angular momentum is $j_B = l_B + s_B = 4$ and g factor $g_B = 5/4$. The magnetization of the system $g_A M^A + g_B M^B$ as a function of the temperature is depicted in figure 1 for parameters $\kappa_A/\kappa = 0.65$ and $\kappa_B/\kappa = 0.0001$.

The figure is in very good agreement with the experimental ZFC magnetization curves [2, 3] (see figure 1 [11]). The parameters κ_A/κ and κ_B/κ are chosen so

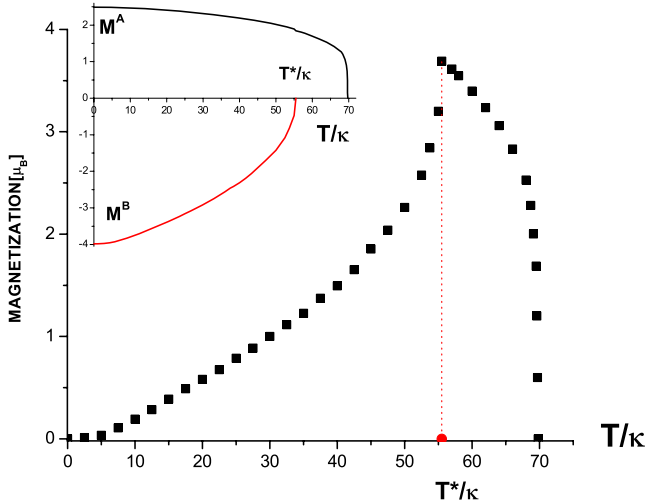


Figure 1. The magnetization $g_A M^A + g_B M^B$ as a function of T/κ for parameters $\kappa_A/\kappa = 0.65$ and $\kappa_B/\kappa = 0.0001$. Inset: the sublattice A (Mn) and B (V) spontaneous magnetization as a function of T/κ . T^* is the temperature at which the spontaneous magnetization of the vanadium ions become equal to zero. (This figure is in colour only in the electronic version)

that the theoretically predicted ratio T_N/T^* is close to the experimental one. The anomalous temperature dependence of the magnetization is reproduced, but there is an important difference between the interpretation of the experimental results in [1–5] and the present theoretical results. In the experimental papers T_N is the temperature at which both the Mn and V magnetization become equal to zero. The modified spin-wave theory predicts two phases: at low temperatures ($0, T^*$) sublattice Mn magnetization and sublattice V magnetization contribute to the magnetization of the system, while at high temperatures (T^*, T_N) only Mn ions have non-zero spontaneous magnetization. The vanadium sublattice magnetization sets in at T^* and evidence for this is the abrupt decrease of magnetization below T^* , which also indicates that the magnetic order of vanadium electrons is antiparallel to the order of Mn electrons.

Two ferromagnetic phases were theoretically predicted, very recently, in spin-fermion systems, which obtain their magnetic properties from a system of localized magnetic moments being coupled to conducting electrons [10]. At the characteristic temperature T^* , the magnetization of itinerant electrons becomes zero, and a high temperature ferromagnetic phase ($T^* < T < T_C$) is a phase where only localized electrons have contributions to the magnetization of the system. An anomalous increasing of magnetization below T^* is obtained, in good agreement with experimental measurements of the ferromagnetic phase of UGe₂ [11].

The results of the present paper and the previous one [10] suggest that the T^* transition from a magnetic phase to another magnetic phase is a generic feature of the two magnetic orders systems. The additional phase transition demonstrates itself through the anomalous temperature variation of the spontaneous magnetization, but it is important to discuss alternative experimental detections of the T^* transition. This

is why we consider the FC magnetization curves [2, 3]. For samples cooled in a field (FC magnetization) the field leads to formation of a single domain and, in addition, increases the chaotic order of the spontaneous magnetization of the vanadium sublattice, which is antiparallel to it. As a result the average value of the vanadium magnetic order decreases and does not compensate for the Mn magnetic order. The magnetization curves depend on the applied field and do not go to zero. For a larger field the (FC) curve increases when the temperature decreases below the Néel temperature. It has a maximum at the same temperature $T^* < T_N$ as the ZFC magnetization and a minimum at $T_1^* < T^*$. Below T_1^* the magnetization increases monotonically when the temperature approaches zero.

The experiments with samples cooled in a field (FC magnetization) provide a new opportunity to clarify the magnetism of the manganese vanadium oxide spinel. The applied field is antiparallel with vanadium magnetic moment and strongly affect it. On the other hand, the experiments show that there is no difference between ZFC and FC magnetization curves when the temperature is over the interval (T^*, T_N) [2, 3]. They begin to diverge when the temperature is below T^* . This is in accordance with the theoretical prediction that the vanadium magnetic moment does not contribute to the magnetization when $T > T^*$, and T^* is the temperature at which the vanadium ions start to form magnetic order. Because of the strong field, the two vanadium bands are split and the magnetic moment of one of the t_{2g} electrons is reoriented to be parallel with the field and magnetic order of Mn electrons. The description of this case is more complicated and requires three magnetic orders to be involved. When $T^* < T < T_N$ only Mn ions have non-zero spontaneous magnetization. At T^* vanadium magnetic order antiparallel with the magnetic order of Mn sets in and partially compensates it. Below T_1^* the reoriented electron gives a contribution, which explains the increasing of the magnetization of the system when the temperature approaches zero. A series of experiments with different applied fields could be decisive for the confirmation or rejection of the T^* transition. Increasing the applied field one expects an increase of T_1^* and, when the field is strong enough, so that all vanadium electrons are reoriented, an anomalous increasing of magnetization below T^* would be obtained as within the ferromagnetic phase of UGe₂ [11].

To conclude, we note that, to account more accurately for the canted magnetic order of the vanadium ions, one has to consider a model (1) with sublattice B exchange constant κ_B depending on space directions: κ_B^z positive (ferromagnetic) and κ_B^x and κ_B^y negative (antiferromagnetic). With appropriately chosen parameters the new model will reproduce the geometry of the vanadium magnetic moment and the main result of the present paper, namely the existence of partial ordering.

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