Antiferromagnetic resonance in transition-metal fluorides

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We evaluate the first quantum correction to the classical expression for the AFMR frequency in transition metal fluorides with a CoF_2 type magnetic structure in the magnetic fields lower than the magnetic sublattices reversing field. Taking the first quantum correction into account leads to a change in the character itself of the functional dependence of the AFMR frequency on the constants in the magnetic Hamiltonian.

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Antiferromagnetic resonance (AFMR) in divalent fluorides of transition metals, in particular, CoF_2,[1] has recently been widely studied. Usually one obtains an expression for the AFMR frequency from Landau-Lifshitz type equations.[2] Oguchi[3] was the first to show that the expression for the AFMR frequency obtained in that way is valid only for large values of the spin S of the ion at a lattice site. The spins of transition metal ions are not very large, e.g., the spin of the cobalt ion equals 3/2 and, hence, quantum corrections to the AFMR spectrum may be appreciable. Oguchi[3] calculated the first quantum correction to the AFMR spectrum and to the expressions for physical quantities for the case of an ideal antiferromagnetic. It turned out that taking quantum corrections into account leaves all physical conclusions of the theory in force. For instance, notwithstanding that in the approximation which goes beyond the classical one the average value of the z-component of the ion spin does not equal S, in uniaxial crystals with an anisotropy of the form

$$-a\sum_a\left[(S_{az}^1)^2+(S_{az}^2)^2\right],$$

S^k_{am} is the m-th component of the spin at site a in the k-th sublattice, the principal conclusion of the theory is retained that the longitudinal magnetic susceptibility $\chi_{\parallel}$ vanishes at zero temperature.[3] This is a consequence of the fact that this kind of anisotropy conserves the z-component of the total spin of the crystal. There exist, however, uniaxial crystals with an anisotropy energy which contains a contribution of a different form. For instance, in the antiferromagnetics $MnCO_3$, $CoCO_3$, and $FeCO_3$ there is an anisotropy[4]

$$\mathscr{H}_D=d\sum_{a,b}(S_{ax}^1S_{by}^2-S_{ay}^1S_{bx}^2).$$

This anisotropy also conserves the z-component of the total spin and $\chi_{\parallel}=0$ at zero temperature. In crystals of transition metal fluorides, e.g., in CoF_2, experiments give $\chi_{\parallel}\neq 0$ at zero temperature.[5] This is connected with the fact that in transition metal fluorides we have an anisotropy of the form[6]

$$\mathscr{H}_D=d\sum_a\left[S_{ax}^1S_{ay}^1+S_{ay}^1S_{ax}^1-S_{ax}^2S_{ay}^2-S_{ay}^2S_{ax}^2\right].$$

This anisotropy does not conserve the z-component of the total spin of the crystal. This also leads to a nonvanishing longitudinal susceptibility at zero temperature. The non-conservation of the z-component of the total spin of the crystal gives particularly favorable conditions for the occurrence of quantum effects in the AFMR spectrum. In the present paper we evaluate the first quantum correction to the AFMR spectrum in transition metal fluorides with a CoF_2 type magnetic structure in magnetic fields smaller than the sublattice reversing field.

Up to second order terms the magnetic Hamiltonian has the form

$$\begin{aligned}\mathscr{H}=&\frac{1}{2}\sum_{a,b}\bar{J}_{ab}(\mathbf{S}_a^1\mathbf{S}_b^1+\mathbf{S}_a^2\mathbf{S}_b^2)+\sum_{a,b}J_{ab}\mathbf{S}_a^1\mathbf{S}_b^2\\&-\frac{a}{2}\sum_a\left[(S_{az}^1)^2+(S_{az}^2)^2\right]+d\sum_a\bigl[S_{ax}^1S_{ay}^1+S_{ay}^1S_{ax}^1\\&-S_{ax}^2S_{ay}^2-S_{ay}^2S_{ax}^2\bigr]+\mu_BgH\sum_a(S_{az}^1+S_{az}^2).\end{aligned}$$

Here $\bar{J}_{ab}$ is the exchange integral corresponding to atoms of one magnetic sublattice, J_{ab} the exchange integral corresponding to atoms of different magnetic sublattices, a and d single-ion anisotropy constants, μ_B the Bohr magneton, $g\equiv g_{zz}$ a component of the g tensor, and H the magnetic field.

The Holstein-Primakoff method[7] enables us to expand the Hamiltonian in powers of 1/S. In the case of an antiferromagnetic it is necessary to introduce two different definitions of spin deviation operators.[3] Expanding the spin operators in powers of 1/S and restricting ourselves to the first two terms, we get

$$\begin{gathered}S_{a-}^1=(2S)^{1/2}\left(1-\frac{a_a^+a_a}{4S}\right)a_a,\quad S_{a+}^2=(2S)^{1/2}\left(1-\frac{b_a^+b_a}{4S}\right)b_a,\\S_{a+}^1=a_a^+(2S)^{1/2}\left(1-\frac{a_a^+a_a}{4S}\right),\quad S_{a-}^2=b_a^+(2S)^{1/2}\left(1-\frac{b_a^+b_a}{4S}\right),\\S_{az}^1=-S+a_a^+a_a,\qquad S_{az}^2=S-b_a^+b_a.\end{gathered}$$

Up to terms of order S^0 the magnetic Hamiltonian expressed in terms of the Fourier components of the spin deviation operators has the form

$$\begin{aligned}\mathscr{H}=&NS^2(\bar{J}_0-J_0-a)+\sum_{\mathbf{k}}\Bigl\{\Bigl[S(-\bar{J}_0+\bar{J}_{\mathbf{k}}+J_0)\\&+a\left(S-\frac{1}{2}\right)+h\Bigr]a_{\mathbf{k}}^+a_{\mathbf{k}}+\Bigl[S(-\bar{J}_0+\bar{J}_{\mathbf{k}}+J_0)\\&+a\left(S-\frac{1}{2}\right)-h\Bigr]b_{\mathbf{k}}^+b_{\mathbf{k}}\Bigr\}+S\sum_{\mathbf{k}}J_{\mathbf{k}}(a_{\mathbf{k}}^+b_{-\mathbf{k}}^+\\&+a_{\mathbf{k}}b_{-\mathbf{k}})+\frac{d}{i}\left(S-\frac{1}{4}\right)\sum_{\mathbf{k}}(a_{\mathbf{k}}^+a_{-\mathbf{k}}^+-a_{\mathbf{k}}a_{-\mathbf{k}}\\&+b_{\mathbf{k}}^+b_{-\mathbf{k}}^+-b_{\mathbf{k}}b_{-\mathbf{k}})-\frac{1}{4N}\sum_{1,2,3,4}\delta(1+2-3-4)(\bar{J}_1\\&+\bar{J}_4-2\bar{J}_{1-4}+2a)(a_1^+a_2^+a_3a_4+b_1^+b_2^+b_3b_4)\\&-\frac{1}{N}\sum_{1,2,3,4}\delta(1+3-2-4)J_{1-2}a_1^+a_2b_3^+b_4\\&-\frac{1}{4N}\Bigl\{\sum_{1,2,3,4}\delta(1+2+4-3)J_4(a_1^+a_2^+a_3b_4^++b_1^+b_2^+b_3a_4^+)\end{aligned}$$

$$+\sum_{1,2,3,4}\delta(2+3+4-1)J_4(a_1^+a_2a_3b_4+b_1^+b_2b_3a_4)\Big\}$$

$$+i\frac{d}{2N}\Big\{\sum_{1,2,3,4}\delta(1+2+3-4)(a_1^+a_2^+a_3^+a_4+b_1^+b_2^+b_3^+b_4)$$

$$-\sum_{1,2,3,4}\delta(2+3+4-1)(a_1^+a_2a_3a_4+b_1^+b_2b_3b_4)\Big\},$$

$\bar J_{\mathbf k}$ is the Fourier component of the exchange integral $\bar J_{ab}$, $J_{\mathbf k}$ the Fourier component of the exchange integral J_{ab}, and $h = \mu_B gH$.

To evaluate the spectrum in the main order in the spin it is necessary to diagonalize that part of the Hamiltonian which is quadratic in the operators. It corresponds to non-interacting spin waves. The diagonalization is realized by the Holstein-Primakoff[7] u, v transformation which couples boson creation and annihilation operators:

$$\begin{aligned} a_{\mathbf k}&=p_{\mathbf k}\alpha_{\mathbf k}+iq_{\mathbf k}\alpha_{-\mathbf k}^{+}+ir_{\mathbf k}\beta_{\mathbf k}-t_{\mathbf k}\beta_{-\mathbf k}^{+},\\ b_{\mathbf k}&=iu_{\mathbf k}\alpha_{\mathbf k}-v_{\mathbf k}\alpha_{-\mathbf k}^{+}+w_{\mathbf k}\beta_{\mathbf k}+ix_{\mathbf k}\beta_{-\mathbf k}^{+}, \end{aligned}\qquad (1)$$

where $p_{\mathbf k}$, $q_{\mathbf k}$, $r_{\mathbf k}$, $t_{\mathbf k}$, $u_{\mathbf k}$, $v_{\mathbf k}$, $w_{\mathbf k}$, and $x_{\mathbf k}$ are real functions. The symmetry group of the crystals considered contains the inversion so that $\bar J_{\mathbf k} = \bar J_{-\mathbf k}$, $J_{\mathbf k} = J_{-\mathbf k}$. We therefore consider a canonical transformation with coefficients which are even functions of the wavevector. As a result of the diagonalization we get the following expression for the magnon dispersion law:[8]

$$\omega_{1,2}(\mathbf k)=[([A_{\mathbf k}^2-B_{\mathbf k}^2]^{1/2}\pm h)^2-C^2]^{1/2};$$

$$A_{\mathbf k}=S(\bar J_{\mathbf k}-\bar J_0+J_0+a),\quad B_{\mathbf k}=SJ_{\mathbf k},\quad h=\mu_B g\mathrm{H},\quad C=2Sd.$$

Restricting ourselves to nearest neighbors we have

$$J_{\mathbf k}=J\sum e^{i\mathbf{ka}},\ \bar J_{\mathbf k}=\bar J\sum e^{i\mathbf{ka}},$$

where J is the exchange integral for atoms, one of which is at the center and the other at the vertex of a cell, $\bar J$ the exchange integral for atoms in neighboring vertexes;

$$J_{\mathbf k}=8J\gamma_{\mathbf k},\quad \gamma_{\mathbf k}=\cos{}^1\!/_2k_x\cos{}^1\!/_2k_y\cos{}^1\!/_2k_z,$$

$$\bar J_{\mathbf k}=6\bar J\bar\gamma_{\mathbf k},\quad \bar\gamma_{\mathbf k}={}^1\!/_3(\cos k_x+\cos k_y+\cos k_z).$$

The wavevector components are here measured in reciprocal lattice lengths.

For H = 0 the expression for the square of the AFMR frequency is

$$\omega^2(0)=S^2(16Ja+a^2)-4S^2d^2.$$

The condition for the stability of the magnetic structure considered, when there is no external magnetic field, is

$$a+a^2/16J-d^2/4J>0$$

in agreement with Dzyaloshinskiĭ.[6] When H ≠ 0 the expression for the square of the AFMR frequencies is

$$\omega_{1,2}^2(0)=(S(16Ja+a^2)^{1/2}\pm h)^2-4S^2d^2$$

and the condition for stability in an external magnetic field is thus

$$S(16Ja+a^2)^{1/2}-2Sd>h.$$

In the approximation considered the ground state energy is

$$E_0=NS(S+1)(\bar J_0-J_0-a)+\frac{1}{2}\sum_{\mathbf k}(\omega_1(\mathbf k)+\omega_2(\mathbf k)),$$

and the magnetic susceptibility along the field

$$\chi_\parallel=-\frac{\partial^2E_0}{\partial H^2}=\frac{(\mu_Bg)^2C^2}{2N}\sum_{\mathbf k}\Big(\frac{1}{\omega_1^3(\mathbf k)}+\frac{1}{\omega_2^3(\mathbf k)}\Big).$$

For the case of a small anisotropy energy, when a, d ≪ J, the expression for the longitudinal susceptibility is

$$\chi_\parallel=\frac{(g\mu_B)^2d^2}{2^{10}\pi^2SJ^{3/2}(J+|\bar J|)^{3/2}}\Big\{\Big(1+\frac{h}{4S(Ja)^{1/2}}\Big)^{-3/2}$$

$$\times\ln\Big[\frac{2^8\pi^2S^2J(J+|\bar J|)(1+h/4S(Ja)^{1/2})}{(4S(Ja)^{1/2}+h)^2-4S^2d^2}\Big]$$

$$+\Big(1-\frac{h}{4S(Ja)^{1/2}}\Big)^{-3/2}\ln\Big[\frac{2^8\pi^2S^2J(J+|\bar J|)(1-h/4S(Ja)^{1/2})}{(4S(Ja)^{1/2}-h)^2-4S^2d^2}\Big]\Big\},$$

we have used here the fact that $\bar J < 0$.

If the anisotropy energy is of the order of the exchange energy the logarithms turn out to be of order unity. We can in that case write down an expression for the longitudinal susceptibility in fields of the order of the reversing field, when the condition

$$1-2d(16Ja+a^2)^{-1/2}-hS^{-1}(16Ja+a^2)^{-1/2}\ll1$$

is satisfied. The main contribution to the susceptibility is

$$\chi_\parallel=\frac{(\mu_Bg)^2d^2}{16\pi^2S}(16Ja+a^2)^{-3/2}(J+|\bar J|)^{-3/2}\Big(1-\frac{h}{S}(16Ja+a^2)^{-1/2}\Big)^{-3/2}\ln\Phi,$$

$$\Phi=\frac{8\pi^2(J+|\bar J|)}{a(1-2d(16Ja+a^2)^{-1/2}-hS^{-1}(16Ja+a^2)^{-1/2})}$$

When $h = S(16Ja + a^2)^{1/2} - 2Sd$ the susceptibility $\chi_\parallel$ becomes infinite in agreement with the fact that the magnetic structure becomes unstable.

The magnetic Hamiltonian written down up to terms of order S^0 contains fourth order terms in the Fourier components of the spin deviation operators. The presence of fourth order terms in the Hamiltonian corresponds to the interaction of the spin waves. The transformation (1) connects boson creation and annihilation operators so that as a result of the transformation of the fourth-order terms in the Hamiltonian we get fourth order-terms in the magnon operators which are not of the form of N-products. After the transformation all terms in the Hamiltonian must be brought to the form of N-products. As a result of bringing the fourth order terms in the magnon operators to the N-product form we get terms which do not contain magnon operators and terms quadratic in the magnon operators. Terms which do not contain magnon operators give a correction of order S^0 to the ground state energy. The coefficients of the terms quadratic in the operators of the form $\alpha^+\alpha$ and $\beta^+\beta$ give a correction $\sim S^0$ to the magnon frequencies, and the coefficients of the terms of the form $\alpha\alpha$, $\alpha^+\alpha^+$, $\beta\beta$, $\beta^+\beta^+$, $\alpha\beta$, $\alpha^+\beta^+$, $\alpha\beta^+$, and $\alpha^+\beta$ corrections of order S^0 to the diagonalization condition. The diagonalization condition is in this approximation a non-linear integral equation. As before, the functions $p_{\mathbf k}$, $q_{\mathbf k}$, $r_{\mathbf k}$, $t_{\mathbf k}$, $u_{\mathbf k}$, $v_{\mathbf k}$, $w_{\mathbf k}$, and $x_{\mathbf k}$ can be chosen to be real.

In the theory of antiferromagnetism it is not the magnon frequency itself, but its square which is expanded in powers of 1/S.[3] It is rather cumbersome to evaluate the contribution, linear in S, to the square of the magnon frequency. We give here only the expression for the square of the AFMR frequencies. For the case of small anisotropy energies, when J ≫ a, d, we have

$$\tilde\omega_{1,2}^2(\mathbf k=0)=16S^2Ja+h^2-4S^2d^2\pm8S(Ja)^{1/2}h+16SJa\gamma$$

$$-4Sd^2(\varepsilon-\delta)\pm4(Ja)^{1/2}h\gamma\pm d^2h(Ja)^{-1/2}\delta.$$

We have dropped in the expression for the square of the AFMR frequencies terms which have ∝ of order a^2 in comparison with terms ∝ Ja. Moreover, we have used the notation

$$\gamma=1-\frac{1}{N}\sum_{\mathbf k}\frac{A_{\mathbf k}^{(0)}}{\Omega_{\mathbf k}^{(0)}}-\frac{1}{2N}\sum_{\mathbf k}\frac{A_{\mathbf k}^{(0)}-\gamma_{\mathbf k}B_{\mathbf k}}{\Omega_{\mathbf k}^{(0)}},$$

$$\delta=\frac{SJ_0}{N}\sum_{\mathbf{k}}\frac{1}{\Omega_{\mathbf{k}}^{(0)}},\quad \varepsilon=1-\frac{2}{N}\sum_{\mathbf{k}}\frac{A_{\mathbf{k}}^{(0)}}{\Omega_{\mathbf{k}}^{(0)}};$$

$$A_{\mathbf{k}}^{(0)}=-\bar{J}_0+\bar{J}_{\mathbf{k}}+J_0,\quad \Omega_{\mathbf{k}}^{(0)}=[A_{\mathbf{k}}^{(0)2}-B_{\mathbf{k}}^2]^{1/2}$$

The contribution to the square of the AFMR frequencies which is linear in S contains two kinds of terms. Terms of the first group in their dependence on the constants of the magnetic Hamiltonian have their counterparts among terms $\propto S^2$. Moreover, there is also a contribution $\pm d^2h(Ja)^{-1/2}\delta$ which has not got an analogy among the terms of order S^2. In the case considered the evaluation of the first quantum correction to the classical expression for the AFMR frequencies has thus led to a change in the character itself of their functional dependence on the constants in the magnetic Hamiltonian. This fact is a consequence of the fact that the z-component of the total spin of the crystal is not conserved.

The spin-orbit interaction is large in CoF_2 and the uni-axial anisotropy constants a and d turn out to be of the order of the exchange integrals J and $\bar{J}$. It has therefore sense to give the complete expression for the AFMR frequencies:

$$\tilde{\omega}_{1,2}^2=\left[\left\{\left[\frac{1}{2}(\tilde{A}+\tilde{A}')-\operatorname{th}(\tilde{\varphi}-\tilde{\chi})C\right]^2-\tilde{B}^2\right\}^{1/2}\pm\left[h+\frac{1}{2}(\tilde{A}-\tilde{A}')\right]\right]^2$$

$$-\left[\mp[A_0^2-B_0^2]^{1/2}\tilde{D}/B_0+\frac{1}{2}(\tilde{C}+\tilde{C}')-\operatorname{th}(\tilde{\varphi}-\tilde{\chi})A_0\right]^2;$$

$$\tilde{A}+\tilde{A}'=(2S+1)(J_0+a)-\frac{1}{2N}\sum_{\mathbf{k}}[(J_0+2a)\operatorname{ch}2\theta_{\mathbf{k}}-J_{\mathbf{k}}\operatorname{sh}2\theta_{\mathbf{k}}]$$

$$\times(\operatorname{ch}2\varphi_{\mathbf{k}}+\operatorname{ch}2\psi_{\mathbf{k}})+\frac{3d}{2N}\sum_{\mathbf{k}}(\operatorname{sh}2\varphi_{\mathbf{k}}+\operatorname{sh}2\psi_{\mathbf{k}}),$$

$$\tilde{A}-\tilde{A}'=(J_0-2a)\frac{1}{2N}\sum_{\mathbf{k}}(\operatorname{ch}2\varphi_{\mathbf{k}}-\operatorname{ch}2\psi_{\mathbf{k}})+\frac{3d}{2N}\sum_{\mathbf{k}}\operatorname{ch}2\theta_{\mathbf{k}}(\operatorname{sh}2\varphi_{\mathbf{k}}-\operatorname{sh}2\psi_{\mathbf{k}}),$$

$$\tilde{B}=\left(S+\frac{1}{2}\right)J_0-J_0\frac{1}{4N}\sum_{\mathbf{k}}(\operatorname{ch}2\theta_{\mathbf{k}}-\gamma_{\mathbf{k}}\operatorname{sh}2\theta_{\mathbf{k}})(\operatorname{ch}2\varphi_{\mathbf{k}}+\operatorname{ch}2\psi_{\mathbf{k}}),$$

$$\tilde{C}+\tilde{C}'=2(2S+1)d+\frac{1}{4N}\sum_{\mathbf{k}}[\bar{J}_0(1-\bar{\gamma}_{\mathbf{k}})+2a](\operatorname{sh}2\varphi_{\mathbf{k}}+\operatorname{sh}2\psi_{\mathbf{k}})$$

$$-\frac{3d}{2N}\sum_{\mathbf{k}}\operatorname{ch}2\theta_{\mathbf{k}}(\operatorname{ch}2\varphi_{\mathbf{k}}+\operatorname{ch}2\psi_{\mathbf{k}}),$$

$$\tilde{D}=\frac{1}{8N}J_0\sum_{\mathbf{k}}(2\gamma_{\mathbf{k}}\operatorname{sh}2\theta_{\mathbf{k}}-\operatorname{ch}2\theta_{\mathbf{k}})(\operatorname{sh}2\varphi_{\mathbf{k}}-\operatorname{sh}2\psi_{\mathbf{k}}),$$

$$\operatorname{th}(\tilde{\varphi}-\tilde{\chi})=\frac{1}{2h}\left\{\frac{1}{4N}\sum_{\mathbf{k}}\left[-\frac{1}{S}(A_{\mathbf{k}}^2-B_{\mathbf{k}}^2)^{1/2}+2a(A_{\mathbf{k}}+\gamma_{\mathbf{k}}B_{\mathbf{k}})(A_{\mathbf{k}}^2-B_{\mathbf{k}}^2)^{-1/2}\right]\right.$$

$$\left.\times(\operatorname{sh}2\varphi_{\mathbf{k}}-\operatorname{sh}2\psi_{\mathbf{k}})-\frac{3d}{2N}\sum_{\mathbf{k}}(\operatorname{ch}2\varphi_{\mathbf{k}}-\operatorname{ch}2\psi_{\mathbf{k}})\right\},$$

$$\operatorname{th}2\theta_{\mathbf{k}}=\frac{B_{\mathbf{k}}}{A_{\mathbf{k}}},\quad \operatorname{sh}2\varphi_{\mathbf{k}}=\frac{C}{[A_{\mathbf{k}}^2-B_{\mathbf{k}}^2]^{1/2}+h},\quad \operatorname{sh}2\psi_{\mathbf{k}}=\frac{C}{[A_{\mathbf{k}}^2-B_{\mathbf{k}}^2]^{1/2}-h}.$$

The part of the ground-state energy which depends on the magnetic field does not reduce to the zero-point vibration energy and the expression obtained for the longitudinal susceptibility is unwieldy. We note merely that in contrast to the leading approximation in the spin the susceptibility can no longer be expressed in terms of the AFMR frequency $\tilde{\omega}$.

As the magnitude of the spins of the transition metal ions is small the results obtained have mainly a theoretical value. However, they show that in the case of transition metal fluorides the dependence of the AFMR frequencies on the constants of the magnetic Hamiltonian which is calculated using Landau-Lifshitz type equations is incorrect. The expression for the longitudinal susceptibility, calculated in the leading order in the spin, is also incorrect.

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