# Closed-form description of the magnetization and of the resonant frequencies of antiferromagnets in a magnetic field. $NiF_2$

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We present in closed form a model-independent phenomenological description of the static and dynamic properties of easy-plane tetragonal antiferromagnets with unquenched orbital angular momentum. The results are used to describe the magnetic properties of nickel fluoride. The presence of several $g$ factors ($\gamma$ factors) of exchange origin predicted by the theory for antiferromagnets was experimentally observed. For $NiF_2$ at $T = 4.2$, the difference between them reaches 50%. It is shown that a one-to-one correspondence exists between the dynamic parameters of the two types of phenomenological equations of motion, Lagrangian and nonequilibrium-thermodynamic. The complete set of phenomenological parameters necessary for the construction of the potential and of the equations of motion is determined; these contain a finite number of terms and describe adequately the static and the dynamic properties of nickel fluoride at $T = 4.2$ K. The derived equations of motion and potential are used to calculate the magnetization components and AFMR frequencies for an arbitrary orientation of the magnetic field in the (001) plane. In the entire magnetic-field interval ($H \leq 65$ kOe) for which reliable measurement results of the magnetization are known at present, the experimental and calculated plots of the magnetization and of the AFMR frequencies agree within the limits of the measurement and calculation accuracy.

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Nickel fluoride is one of the most thoroughly experimentally studied antiferromagnets. The interest in $NiF_2$ is due mainly to two cases. On the one hand, to solve the fundamental problems of the physics of antiferromagnetism it is necessary to construct a theory that describes the properties of substances for which the contribution of the orbital magnetism to the sublattice magnetism cannot be neglected. A study of the properties of $NiF_2$ casts light on various monitor stations of the contribution made to the total magnetic moment by the orbital component. On the other hand, $NiF_2$ differs from analogous antiferromagnets in having a relatively small anisotropy compared with the exchange interaction. Therefore, together with the simplicity of the structure, the relative smallness of the effective anisotropy fields makes it possible to use $NiF_2$ as an example for the development of the rigorous phenomenological theory that describes the experiment adequately. The published attempts to contruct a theory suffer either from unjustifiably excessive simplification of the model of the magnetic subsystem, or from the lack of rigor, from the point of view of general principles, in the assumptions made concerning the connection between the kinetic coefficients and the static effective fields. A detailed analysis of the difficulties in the presently existing approaches to the description of the properties of antiferromagnets is contained in Ref. 1.

The purpose of the present investigation is to develop a model-independent phenomenological approach[2–8] for a description, in closed form, of the static and the dynamic properties of antiferromagnets with unquenched orbital angular momentum. For the reasons indicated above, $NiF_2$ was chosen as an example.

In the paramagnetic state, $NiF_2$ has a tetragonal crystal structure of the rutile type (Fig. 1)–space group $D_{4h}^{14} - P4_2/mnm$, $T_N = 73.2$ K. At $T = 4.2$ K and $H = 0$, the magnetic moments of the sublattices $M_1$ and $M_2$ lie in the (001) plane, so that $\mathbf{M} \perp \mathbf{L}$ ($\mathbf{M} = \mathbf{M}_1 + \mathbf{M}_2$, $\mathbf{L} = \mathbf{M}_1 - \mathbf{M}_2$) and $\mathbf{M} \parallel [100]$ (or $\mathbf{M} \parallel [010]$).

The macroscopic phenomenological description of tetragonal antiferromagnets admits, at definite orientations of the magnetic field $\mathbf{H}$ (for example $\mathbf{H} \parallel [110]$) of inequality of the magnetic moments of the sublattices. In other words, the magnetic moments of the sublattices in such crystals depend not only on the temperature and on the magnetic field, but also on the orientations of the moments relative to the crystallographic axes.[9] In the dynamics, the dependence of the magnetic moments of the sublattices on the orientation, upon deviation from the equilibrium position, can be taken into account within the framework of the thermodynamics of nonequilibrium processes[2] and within the framework of Lagrangian spin-wave mechanics,[10,11] which is rigorously valid at $T = 0$ K. Since a one-to-one connection exists at $T = 0$ K between the dynamic phenomenological parameters of the two approaches,[1,8] it is possible to describe AFMR at low temperatures by the equations of either type. However, if we start with the Landau-Lifshitz equations, whose direct generalization are the nonequilibrium-thermodynamics equations, then

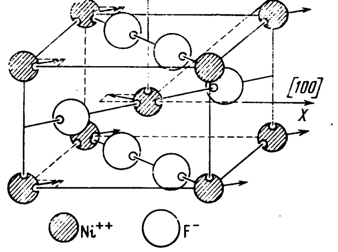


FIG. 1. Magnetic structure of $NiF_2$.


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it turns out that the relative magnitude of the dynamic parameters of the spin-wave Lagrangian mechanics depend substantially on the ground state of the antiferromagnet. We shall therefore use as the basic equations of the dynamics of the magnetic subsystem of $NiF_2$ the thermodynamic equations[2]

$$\begin{pmatrix} \dot{\mathbf{m}} \\ \dot{\mathbf{l}} \end{pmatrix} = \hat{\gamma} \begin{pmatrix} \mathbf{F} \\ \mathbf{N} \end{pmatrix}, \tag{1}$$

where $\mathbf{m}=\mathbf{M}-\mathbf{M}_0$, $\mathbf{l}=\mathbf{L}-\mathbf{L}_0$ are the deviations of the vectors $\mathbf{M}$ and $\mathbf{L}$ from their equilibrium values $\mathbf{M}_0$ and $\mathbf{L}_0$, $\mathbf{F}\equiv\partial\Delta\Phi/\partial\mathbf{m}$ $\mathbf{N}\equiv\partial\Delta\Phi/\partial\mathbf{l}$, $\Delta\Phi$ is the increment to the Landau thermodynamic potential $\Phi$ when the magnetic subsystem deviates from the equilibrium position, and $\hat{\gamma}$ is an antisymmetrical matrix[4]:

$$\begin{pmatrix} 0 & \gamma_{12} & \gamma_{13} & \gamma_{14} & \gamma_{15} & \gamma_{16} \\ & 0 & \gamma_{23} & \gamma_{24} & \gamma_{25} & \gamma_{26} \\ & & 0 & \gamma_{34} & \gamma_{35} & 0 \\ & & & 0 & \gamma_{45} & \gamma_{46} \\ & & & & 0 & \gamma_{56} \\ & & & & & 0 \end{pmatrix}, \tag{2}$$

where

$$\gamma_{12}=-\gamma_1(1+\tau_1)M_{z0},\quad \gamma_{13}=\gamma_1(M_{y0}+\tau_2L_{x0}+\tau_3M_{y0}),$$
$$\gamma_{14}=\gamma_2\tau_4M_{z0},\quad \gamma_{15}=-\gamma_2(1+\tau_5)L_{z0},\quad \gamma_{16}=\gamma_2(L_{y0}+\tau_6M_{x0}+\tau_7L_{y0});$$
$$\gamma_{23}=-\gamma_1(M_{x0}+\tau_2L_{y0}+\tau_3M_{x0}),\quad \gamma_{24}=\gamma_2(1+\tau_5)L_{z0},$$
$$\gamma_{25}=-\gamma_2\tau_4M_{z0},\quad \gamma_{26}=-\gamma_2(L_{x0}+\tau_6M_{y0}+\tau_7L_{x0});$$
$$\gamma_{34}=-\gamma_2(L_{y0}+\tau_8M_{x0}+\tau_9L_{y0}),\quad \gamma_{35}=\gamma_2(L_{x0}+\tau_8M_{y0}+\tau_9L_{x0});$$
$$\gamma_{45}=-\gamma_3(1+\tau_{10})M_{z0},\quad \gamma_{46}=\gamma_3(M_{y0}+\tau_{11}L_{x0}+\tau_{12}M_{y0});$$
$$\gamma_{56}=-\gamma_3(M_{x0}+\tau_{11}L_{y0}+\tau_{12}M_{x0}).$$

The phenomenological parameters $\gamma_1, \gamma_2, \gamma_3$ and $\tau_1, \tau_2, \ldots, \tau_{12}$ are respectively of exchange and anisotropy origin. It should be noted that the region of applicability of Eqs. (1) is limited by the spin-density velocity, which should be low enough to be able to satisfy for the AFMR frequencies the relation $\omega^2 \ll (\gamma H_E)^2$, where $\gamma$ is the gyromagnetic ratio for the magnetic ion and $H_E$ is the interatomic exchange field. When this condition is violated it is necessary to take into account in the equations of motion the second derivative with respect to time, or else the part of the Lagrange function which is bilinear in the velocities in the approach of Dzyaloshinskiĭ and Kukharenko.[10] It is shown in Ref. 1 that to describe the AFMR in $NiF_2$ on the basis of the Lagrangian equations of motion it is necessary to take into account in the phenomenological Lagrangian only the terms that lead to the first derivative with respect to time in the equations of motion. Allowance for the second derivatives leads not only to a renormalization of the dynamic parameters but also the appearance in $NiF_2$ of AFMR frequencies not observable in experiment.

To calculate the AFMR frequencies from the equations of motion (1) it is necessary to determine the equilibrium values of the components of the vectors $\mathbf{M}_0$ and $\mathbf{L}_0$. The values of $\mathbf{M}_0$ and $\mathbf{L}_0$ are the solutions of six nonlinear equations[9]:

$$(\partial\Phi/\partial M_i)_0=H_i,\quad (\partial\Phi/\partial L_j)_0=0,\quad i,j=x,y,z. \tag{3}$$

The complete rational basis of invariants (CRBI) for tetragonal two-sublattice antiferromagnets contains thirteen invariants[1,4,5]

$$I_1=\mathbf{L}^2,\quad I_2=\mathbf{M}^2,\quad I_3=(\mathbf{LM})^2,\quad I_4=L_z^2,\quad I_5=M_z^2,\quad I_6=(\mathbf{LM})L_zM_z,$$
$$I_7=L_xM_y+L_yM_x,\quad I_8=(\mathbf{LM})L_xL_y,\quad I_9=L_xL_yL_zM_z,\quad I_{10}=L_x^2L_y^2,$$
$$I_{11}=(\mathbf{LM})M_xM_y,\quad I_{12}=L_zM_zM_xM_y,\quad I_{13}=M_x^2M_y^2. \tag{4}$$

Assuming the thermodynamic potential $\Phi$ to be a function of the invariants (4), we can write the system (3) in the form

$$\sum_{k=1}^{13}\Phi_k(\partial I_k/\partial M_i)_0=H_i,\qquad \sum_{k=1}^{13}\Phi_k(\partial I_k/\partial L_j)_0=0,\qquad i,j=x,y,z, \tag{5}$$

where

$$\Phi_k\equiv(\partial\Phi/\partial I_k)_0,\ \Phi_k=\Phi_k(I_1, I_2, \ldots, I_{13}).$$

For example, for the case $\mathbf{H}\parallel[010]$, which has been well-investigated experimentally for $NiF_2$, the system (5) becomes

$$2\Phi_2M_{y0}+L_{x0}\Phi_7=H_y,\ 2\Phi_1L_{x0}+M_{y0}\Phi_7=0. \tag{6}$$

To solve the system (6) we use the fact that $M/L \ll 1$ in $NiF_2$ at $H \ll H_E$ and $T=4.2$ K. We therefore seek the solution of (6) in series form

$$M_{y0}=M_y^{(1)}+M_y^{(2)}+\ldots,\qquad L_{x0}=L_x^{(0)}+L_x^{(1)}+L_x^{(2)}+\ldots,$$

where $M_y^{(n+1)}/M_y^{(n)} \sim L_x^{(m+1)}/L_x^{(m)} \sim M/L \ll 1$. To solve Eq. (6) we expand $\Phi$ in a series in the components $\mathbf{M}$ and $\mathbf{L}$, assuming $\Phi_i$ to be functions of the basis invariants (4). Then, leaving out the intermediate steps, which can be found in Ref. 1, we obtain

$$M_y^{(1)}/L_0=(H_y^{(1)}-L_0\tilde{\Phi}_7^{(1)})/2\tilde{\Phi}_2^{(0)}L_0,\qquad L_x^{(1)}=0,$$

where the tilde over the phenomenological parameters $\Phi_i$

$$\frac{M_y^{(2)}}{L_0}=-\frac{(3\tilde{\Phi}_{7,2}^{(0)}L_0^3+{}^1/_2\tilde{\Phi}_{7,7,7}^{(0)}L_0^5)}{2\tilde{\Phi}_2^{(0)}L_0}\left(\frac{M_y^{(1)}}{L_0}\right)^2,$$

$$\frac{L_x^{(2)}}{L_0}=-\frac{(\tilde{\Phi}_7^{(1)}+\tilde{\Phi}_{1,7}^{(1)}2L_0^2)+(M_y^{(1)}/L_0)(2\tilde{\Phi}_{1,2}^{(0)}L_0^2)}{\tilde{\Phi}_{1,1}^{(0)}(2L_0)^2}\left(\frac{M_y^{(1)}}{L_0}\right),$$

means that these parameters are sums of infinite series containing only integer powers of $L_0$ at $M=H=0$, while the superscripts in the parentheses denote the higher powers of $M/L$, to which the ratios of these parameters to $L_0\tilde{\Phi}_2^{(0)}\equiv H_E$ are proportional.

In the calculations of the AFMR frequencies at $\mathbf{H}\parallel[010]$ it is convenient to change over the variables $\mathbf{x}\equiv\{m_x, l_y, m_z, m_y, l_x, l_z\}$. Then, recognizing that at $\mathbf{H}\parallel[010]$ we have $\mathbf{M}_0=\{0, M_{y0}, 0\}$, $\mathbf{L}_0=\{L_{x0}, 0, 0\}$, the nonzero elements of the antisymmetrical matrix $\hat{\gamma}$ are

$$\gamma_{13}=\gamma_1[M_{y0}(1+\tau_3)+\tau_2L_{x0}],\quad \gamma_{23}=-\gamma_2[L_{x0}(1+\tau_9)+\tau_8M_{y0}],$$
$$\gamma_{46}=-\gamma_2[L_{x0}(1+\tau_7)+\tau_6M_{y0}],\ \gamma_{56}=\gamma_3[M_{y0}(1+\tau_{12})+\tau_{11}L_{x0}]. \tag{7}$$

In the calculation of the elements of the matrix of the second derivatives (of the stability matrix) $\alpha_{ik}\equiv(\partial^2\Phi/\partial X_i\partial X_k)_0$ we regard the thermodynamic potential $\Phi$ as a function of thirteen basis invariants (4)

$$\alpha_{ik}=\Phi_{\alpha,\beta}\left(\frac{\partial I_\alpha}{\partial X_i}\right)_0\left(\frac{\partial I_\beta}{\partial X_k}\right)_0+\Phi_\alpha\left(\frac{\partial^2 I_\alpha}{\partial X_i\,\partial X_k}\right)_0\qquad(\alpha,\beta=1,2,\ldots,13).$$

In the considered case $\mathbf{H}\parallel[010]$, the only nonzero elements of the symmetrical stability matrix $\alpha$ are

$$\alpha_{11}=2(\Phi_2+\Phi_3L_{x0}^2),\quad \alpha_{12}=\Phi_7+L_{x0}^2\Phi_8+2\Phi_3L_{x0}M_{y0},$$
$$\alpha_{22}=2\Phi_3M_{y0}^2+2\Phi_1+2L_{x0}M_{y0}\Phi_8+2\Phi_{10}L_{x0}^2,\ \alpha_{33}=2(\Phi_2+\Phi_5),$$
$$\alpha_{44}=2\Phi_2+\Phi_{7,7}L_{x0}^2+\Phi_{2,7}2L_{x0}M_{y0}+\Phi_{2,2}4M_{y0}^2,$$
$$\alpha_{45}=\Phi_7+2\Phi_{1,7}L_{x0}^2+(\Phi_{7,7}+4\Phi_{1,2})L_{x0}M_{y0}+2\Phi_{2,7}M_{y0}^2,$$
$$\alpha_{55}=2\Phi_1+\Phi_{1,1}4L_{x0}^2+\Phi_{7,7}M_{y0}^2,\ \alpha_{66}=2(\Phi_1+\Phi_4). \tag{8}$$

Solving the equation $\det|i\omega\hat{1}-\hat{\gamma}\hat{\alpha}|=0$ with $\hat{\gamma}$ from (7) and $\hat{\alpha}$ from (8), we obtain the following expressions for the high- and low-frequency branches $\omega_1$ and $\omega_2$ of the AFMR in an easy-plane tetragonal antiferromagnet at $\mathbf{H}\|[010]$:

$$\omega_1^2=\alpha_{33}(\alpha_{11}\gamma_{13}^2-2\alpha_{12}\gamma_{13}\gamma_{23}+\gamma_{23}^2\alpha_{22}), \quad (9)$$

$$\omega_2^2=\alpha_{66}(\alpha_{44}\gamma_{46}^2-2\alpha_{45}\gamma_{46}\gamma_{56}+\gamma_{56}^2\alpha_{55}). \quad (10)$$

Since the quantities $\gamma_{ik}$ and $\alpha_{jl}$ are rather complicated functions of $\mathbf{M}_0$ and $\mathbf{L}_0$, which in turn are the solutions of the system of equations (6), it may turn out that the orders of magnitude of the various terms in relations (8)–(10) differ. At this stage of the solution it is practically always necessary to take correct account of the orders of smalless in accordance with the actual properties of the real crystal. We point out that a similar situation will always arise in a rigorous and consistent phenomenological description of substances of any symmetry. Of course, it must be borne in mind that if we introduce model-dependent descriptions (thermodynamic potential with a small number of terms, the condition $\mathbf{M}_i^2=$const corresponding to the Landau-Lifshitz equation,[12] and the condition $S_i^2=$const corresponding to the Turov equations[13]), then the relations for the frequencies, even in the case of an arbitrary orientation of the magnetization can simplify substantially. However, these simplifications must be well founded and lead to limitations on both the temperature intervals and on the magnitudes and directions of the magnetic fields at which they can be used. Since no such limitations are imposed in the present paper, we must determine the accuracy with which the phenomenological parameters $\gamma_{ik}$, $\alpha_{jl}$ should be employed in Eqs. (9) and (10). To this end we compare, at $T=4$ K, the AFMR frequencies measured in $NiF_2$ with the exchange frequency $\omega_E$ ($\omega_E=\gamma H_E$), and the measured magnetic moment with the sublattice magnetization $L_0=2M_0=14.400$ cgs emu/mole assumed at $H=0$.[14] At $T=4$ K and $H=0$ we have $\nu_1=4$ cm$^{-1}$,[15] and $=31$ cm$^{-1}$.[16] Putting $\gamma=0.1$ cm$^{-1}$/kOe and $H_E=1200$ kOe,[14] we get $\nu_E=120$ cm$^{-1}$, $\omega_1/\omega_E=3\%$, and $\omega_2/\omega_E=25\%$. The magnetic moment at $H=60$ kOe is $M/L=4\%$ if measured along the [010] axis ($\mathbf{H}\|[010]$) and $M/L=3\%$ along the [110] axis ($\mathbf{H}\|[110]$). From the presented estimates it follows that the expansion of $(\omega_1/\omega_E)^2$ should begin with terms proportional to $(M/L)^2$, while the expansion of $(\omega_2/\omega_E)^2$ should begin with terms proportional to $M/L$. The maximum degree of the expansion is limited by the accuracy of the measurements of $\omega(H)$.

We now describe the method used to obtain from the general equations (9) and (10) for the AFMR frequencies in $NiF_2$ at $\mathbf{H}\|[010]$ the concrete functions $\omega_1^2(H)$ and $\omega_2^2(H)$. We shall show how to take into account in the derivation of the equations for $\omega_1^2(H)$ and $\omega_2^2(H)$ both the measured AFMR frequencies and magnetization itself, and the errors in their measurements. In accordance with the statements made above, we obtain the AFMR frequencies by expanding in the single small parameter $M/L$.

We consider first the equation for the low-frequency branch of the AFMR ($\omega_1$). As indicated above, from the experimental AFMR data[15] we have $(\omega_1/\omega_E)^2\sim(M/L)^2$, i.e., the expansion of each of the three terms of Eq. (9) in powers of the small parameter $M/L$ must contain terms of second, third, fourth, etc. order of smallness. We first take into account only the second-order terms. Then, inasmuch as $(M/L)^2\approx[(H_{D\perp}+H)/H_E]^2$, the right-hand side of Eq. (9) for $\omega_1^2$ will contain terms that are quadratic, linear, and of zero order in the field:

$$\omega_1^2=B_0+B_1H+B_2H^2, \quad (11)$$

and all of these appear when account is taken of terms of second order of smallness and are themselves of the same order of magnitude:

$$B_0/(\gamma H_E)^2\sim B_1H/(\gamma H_E)^2\sim B_2H^2/(\gamma H_E)^2\sim(M/L)^2.$$

If we now take into account the terms of third order of smallness in (9), then the coefficients $B_0, B_1, B_2$ will contain corrections that depend on the magnetic field and are of the order of $M/L$:

$$\omega_1^2=B_0(1+b_{01})+B_1(1+b_{11})H+B_2(1+b_{21})H^2,$$

where $b_{01}(H)\sim b_{11}(H)\sim b_{21}(H)\sim M/L$.

Allowance for the fourth-order terms leads to corrections of order $(M/L)^2$ in $B_0, B_1, B_2$:

$$B_0(1+b_{01}+b_{02}),\ B_1(1+b_{11}+b_{12}),\ B_2(1+b_{21}+b_{22}),$$

where

$$b_{02}(H)\sim b_{12}(H)\sim b_{22}(H)\sim(M/L)^2.$$

Naturally, if we separate in all the indicated corrections to $B_0, B_1, B_2$ the parts that do and do not depend on the magnetic field, then Eq. (9) for $NiF_2$ will contain terms $B_3H^3\sim(M/L)^3$ and $B_4H^4\sim(M/L)^4$, while the corrections to $B_0$, $B_1$, and $B_2$ will no longer depend on the magnetic field.

The AFMR frequencies of $NiF_2$ were measured at $T=4$ K and in fields up to 140 kOe.[14] At $\mathbf{H}\|[010]$ the positions of the absorption lines corresponding to the low-frequency branch of the AFMR were observed in 67 points. At first, all the experimental points were reduced with a computer by least squares using Eq. (11), and the following values of the coefficients were obtained:

$$B_0=12700\pm200\,\text{GHz}^2,\ B_1=935\pm10\ \text{GHz}^2/\text{kOe},\ B_2=10.32\pm0.08\ \text{GHz/kOe})^2. \quad (12)$$

We see that the relative error in the determination of $B_0$, $B_1$, and $B_2$ is less than $M/L$. Consequently, when the right-side of (9) for $NiF_2$ is expanded in powers of the small parameter it is essential to take into account the terms of third order of smallness. There is no need to take into account the fourth-order terms in (9), since they lead to corrections of the order of $(M/L)^2$, meaning an exaggeration of the accuracy with which the coefficients $B_0$, $B_1$, and $B_2$ are determined in (12).[15]

To determine the contribution made by the third-order terms to Eq. (9), we reduced the experimental points by least squares both using the equation

$$\omega_1^2=D_0+D_1H+D_2H^2+D_3H^3, \quad (13)$$

and using Eq. (11), and gradually discarded the points in stronger fields. As a result of the successive discarding of half the points located in strong fields, corresponding to a decrease of the maximum field from 140 to 40 kOe, it was found that the coefficient $B_0$ changes in the range $\pm 0.3\%$, and the variation of the coefficients $B_1$ and $B_2$ is within $\pm 1\%$. Such a "stability" of the values of $B_0, B_1, B_2$ indicates that in $NiF_2$ at $T = 4$ K the contributions of the third-order terms in Eq. (9) for the low-frequency AFMR branch are equal to zero, and the next nonzero contributions can be those of fourth order. In fact, if the third-order terms were different from zero, this would introduce into $B_0, B_1, B_2$ corrections whose values would depend on the maximum field, and if $H_{max}$ were to increase from 40 ($M/L \approx 3\%$) to 140 ($M/L \approx 8\%$) kOe these corrections to $B_0, B_1, B_2$ would amount to $\approx 5\%$.

We have also reduced the indicated experimental points by least squares, using Eq. (13). If all the 67 experimental points are taken into account we have $D_0 = 13480 \pm 260$ GHz$^2$, $D_1 = 910 \pm 22$ GHz$^2$/kOe, $D_2 = 10.7 \pm 0.4$ GHz$^2$/kOe$^2$, and $D_3 = -0.0016 \pm 0.0022$ GHz$^2$/kOe$^2$. It is seen that within the limits of errors the values of $D_i$ and $B_i$ ($i = 1, 2, 0$) are in agreement, and the variance of $D_3$ is practically equal to $D_3$ itself. Let us estimate the "theoretically expected" values of $D_3$. Since $D_3H^3 \sim (M/L)^3$, $D_0 \sim D_1H \sim D_2H^2 \sim (M/L)^2$, it follows that $D_3H^3/D_2H^2 \sim (M/L)$. Consequently, $D_3 \sim (D_2/H)(M/L) \sim 10^{-2}$. It is seen that the experimentally determined value of $D_3$ is smaller by approximately one order of magnitude than the obtained estimates. This favors the conclusion drawn above, that the third-order terms in (9) vanish. Attention must be called, however, to the fact that this conclusion pertains only to those third-order terms which contain $(M/L)^n (n = 1, 2, 3)$ as factors, i.e., depend on the magnetic field. Equation (9) contains in addition third-order terms that do not have $(M/L)^n$ ($n = 1, 2, 3$) as factors, i.e., do not depend on $H$. Third-order terms of this type are contained only in $B_0$. For this reason the contribution made to $B_0$ by the indicated third-order terms cannot be determined by the procedures cited above for the reduction of the experimental points, which are based on the determination of the contribution of different powers of $H$ to the $\omega(H)$ dependence.

Thus, on the basis of the analysis of the experimental data we should retain in the right-hand side of Eq. (9) only terms of second order of smallness in $M/L$ for the low-frequency branch of the AFMR in $NiF_2$, in accordance with the existing experimental results, while the third-order terms, which are contained as the coefficients of $(M/L)^n$ ($n = 1, 2, 3$), should be set equal to zero. Analysis[1] of all three terms in the right-hand side shows that the equation for the low-frequency AFMR branch in $NiF_2$ at $\mathbf{H} \parallel [010]$, accurate to terms of second order of smallness, is

$$\omega_1^2 = \alpha_{33}^{(0)} [\alpha_{11}^{(0)} \tilde{\gamma}_{13}^{(1)2} - 2\tilde{\gamma}_{13}^{(1)} \tilde{\gamma}_{23}^{(0)} \alpha_{12}^{(1)} + \gamma_{23}^{(0)2} \alpha_{22}^{(2)}], \tag{14}$$

where

$$\alpha_{33}^{(0)} = 2\tilde{\Phi}_2^{(0)} \qquad \alpha_{11}^{(0)} = 2(\tilde{\Phi}_2^{(0)} + \tilde{\Phi}_3^{(0)} L_0^2), \qquad \gamma_{13}^{(1)} = \gamma_1 [M_y^{(1)} (1 + \tilde{\tau}_3^{(0)})],$$

$$\gamma_{23}^{(0)} = \gamma_2 L_0 (1 + \tilde{\tau}_9^{(0)}), \qquad \alpha_{22}^{(2)} = 2\tilde{\Phi}_3^{(0)} M_y^{(1)2} - \tilde{\Phi}_7^{(1)} M_y^{(1)}/L_0$$
$$+ 2L_0 \tilde{\Phi}_8^{(1)} M_y^{(1)} + 2L_0^2 \tilde{\Phi}_{10}^{(2)}.$$

We proceed next to an examination of Eq. (10) for the high-frequency branch of the AFMR in $NiF_2$ at $\mathbf{H} \parallel [010]$. As indicated above, it follows from the measurements of Ref. 16 that $(\omega_2/\omega_E)^2 \sim (M/L)$. Therefore the equation for $\omega_2^2$ should contain terms of first and of higher orders of smallness. The maximum order of smallness of the terms in the expansion of $\omega_2^2$ is limited by the accuracy of the measurements. It follows from the results obtained by Richards[16] that $\nu_2 = c_0 + c_1 H$, i.e., $\nu_2^2 = c_0^2 + 2c_0c_1H + c_1^2H^2$, where $c_0 = 31.14 \pm 0.01$ cm$^{-1}$, and $c_1 = 0.0045 \pm 0.0015$ cm$^{-1}$/kOe. In fields $\sim 50$ kOe, the strongest used for the measurements in Ref. 16,

$$\nu_2^2 = c_0^2 [(1 \pm 0.006) + 0.014(1 \pm 0.3) + 5 \cdot 10^{-5} (1 \pm 0.6)].$$

It is seen that in accord with the presently attainable accuracy[16] it is useful to retain in (10), besides the first-order terms, only the terms of second order of smallness, which are nonlinear in the field (in $M$). The second-order terms quadratic in the magnetic field (in $M$) should be set equal to zero. Terms of third and higher order of smallness need not be taken into account, since they are smaller than the measurement errors. Leaving out the analysis of Eq. (10), which is given in Ref. 1, we obtain for $\omega_2^2$ an equation in which, in accord with the experimental results,[16] account is taken only of terms of first and second order of smallness:

$$\omega_2^2 = \alpha_{66}^{(1)} \alpha_{44}^{(0)} \gamma_{46}^{(0)2} + \alpha_{66}^{(1)} \alpha_{55}^{(0)} \gamma_{56}^{(0)2} + \alpha_{66}^{(2)} \alpha_{44}^{(0)} \gamma_{46}^{(0)2}$$
$$+ \alpha_{66}^{(1)} \alpha_{44}^{(0)} (\gamma_{46}^2)^{(1)} + \alpha_{66}^{(1)} \alpha_{44}^{(1)} (\gamma_{46}^{(0)})^2 - 2\alpha_{66}^{(1)} \alpha_{45}^{(1)} \gamma_{46}^{(0)} \gamma_{56}^{(0)} + \alpha_{66}^{(1)} \alpha_{55}^{(0)} (\gamma_{56}^2)^{(1)}, \tag{15}$$

where

$$\alpha_{66}^{(1)} = 2\tilde{\Phi}_4^{(1)}, \quad \alpha_{66}^{(2)} = -\tilde{\Phi}_7^{(1)} M_y^{(1)}/L_0, \quad \alpha_{44}^{(0)} = 2\tilde{\Phi}_2^{(0)}, \quad \alpha_{44}^{(1)} = \tilde{\Phi}_{7,7}^{(1)} L_0^2,$$
$$\alpha_{45}^{(1)} = \tilde{\Phi}_7^{(1)} + 2\tilde{\Phi}_{1,7}^{(1)} L_0^2 + 4\tilde{\Phi}_{1,2}^{(0)} L_0 M_y^{(1)}, \quad \alpha_{55}^{(0)} = 4L_0^2 \tilde{\Phi}_{1,1}^{(0)},$$
$$\gamma_{46}^{(0)} = \gamma_2 L_0 (1 + \tilde{\tau}_7^{(0)}), \quad \gamma_{56}^{(0)} = \gamma_3 \tilde{\tau}_{11}^{(0)} L_0, \quad (\gamma_{46}^2)^{(1)} = \gamma_2^2 2L_0 (1 + \tilde{\tau}_7^{(0)}) \tilde{\tau}_6^{(0)} M_y^{(1)},$$
$$(\gamma_{56}^2)^{(1)} = \gamma_3^2 2M_y (1 + \tilde{\tau}_{12}) \tilde{\tau}_{11} L_0.$$

It is seen from (15) that the formula for the high-frequency AFMR branch for $NiF_2$ at $\mathbf{H} \parallel [010]$, written out accurate to terms of second order of smallness, contains six dyanmic phenomenological parameters, of which four ($\tilde{\tau}_6^{(0)}, \tilde{\tau}_7^{(0)}, \tilde{\tau}_{11}^{(0)}, \tilde{\tau}_{12}^{(0)}$) are of anisotropy origin and two ($\tilde{\gamma}_2, \tilde{\gamma}_3$) are of exchange origin.

In the determination of the orders of $\alpha_{ik}$ and $\gamma_{jl}$ in (9) and (10) it was assumed that the anisotropic dynamic phenomenological parameters $\tilde{\tau}_3, \tilde{\tau}_6, \tilde{\tau}_7, \tilde{\tau}_9, \tilde{\tau}_{11}, \tilde{\tau}_{12} \sim 1$, i.e., they are of zero order of smallness. If, for example, we consider separately one equation of (14) for the low-frequency AFMR branch, then allowance for $\tilde{\tau}_3^{(0)}$ and $\tilde{\tau}_9^{(0)}$ does not lead to an increase of the number of dynamic parameters, inasmuch as after a simple renormalization

$$\tilde{\gamma}_1 (1 + \tilde{\tau}_3^{(0)}) \to \tilde{\gamma}_1', \quad \tilde{\tau}_2^{(0)}/(1 + \tilde{\tau}_3^{(0)}) \to \tilde{\tau}_2'^{(0)}, \quad \tilde{\gamma}_2 (1 + \tilde{\tau}_9^{(0)}) \to \tilde{\gamma}_2'.$$

Eq. (14) will contain only three dynamic parameters ($\tilde{\gamma}_1', \tilde{\gamma}_2', \tilde{\tau}_2'^{(0)}$) and will take the same form as when $\tilde{\tau}_3 = \tilde{\tau}_9 = 0$. However, when the low-frequency ($\omega_1$) and high-frequency ($\omega_2$) AFMR branches are described in closed form, Eq. (14) for $\omega_1$ contains $\tilde{\gamma}_2(1 + \tilde{\tau}_9^{(0)})$, while Eq. (15) for $\omega_2$ contains $\gamma_2(1 + \tilde{\tau}_7^{(0)})$. Therefore a single renormalization for the two branches is impossible, and consequently the ratio of the $\gamma$ factors determined from Eqs. (14) and (15) for the two AFMR branches should equal $\tilde{\gamma}_2(1+\tilde{\tau}_9^{(0)})/\tilde{\gamma}_2(1+\tilde{\tau}_7^{(0)})$. The magnitude (order of smallness) of $\tilde{\tau}_7$ or $\tilde{\tau}_9$ can be determined within the framework of the phenomenological approach only experimentally. If it turns out that the $\gamma$ factors determined from the measurements of both branches differ by less than $M/L \approx 3-4\%$, then we can say that $\tilde{\tau}_7$ and $\tilde{\tau}_9$ are at least of first order of smallness ($\tilde{\tau}_7^{(1)}$, $\tilde{\tau}_9^{(1)}$). In this case $\tilde{\tau}_9^{(1)}$ will enter in the third-order terms of Eq. (14) for the low-frequency AFMR branch, while $\tilde{\tau}_7^{(1)}$ will enter in the second-order terms of Eq. (15) for the high-frequency branch. If we assume that $\tau_3$, $\tau_6$, $\tilde{\tau}_7$, $\tilde{\tau}_9$, $\tilde{\tau}_{11}$ are not of zeroth but of first order of smallness, then

$$\omega_1^2=\overset{(0)}{\alpha_{33}}[\overset{(0)}{\alpha_{11}}(\overset{(1)}{\gamma_{13}})^2-2\overset{(1)}{\gamma_{13}}\overset{(0)}{\gamma_{23}}\overset{(1)}{\alpha_{12}}+\overset{(0)2}{\gamma_{23}}\overset{(2)}{\alpha_{22}}], \tag{16}$$

$$\omega_2^2=\overset{(1)}{\alpha_{66}}\overset{(0)}{\alpha_{44}}\overset{(0)2}{\gamma_{46}}+[\overset{(2)}{\alpha_{66}}\overset{(0)}{\alpha_{44}}\overset{(0)2}{\gamma_{46}}+\overset{(1)}{\alpha_{66}}\overset{(0)}{\alpha_{44}}(\gamma_{46}^2)^{(1)}+\overset{(1)}{\alpha_{66}}\overset{(1)}{\alpha_{44}}\overset{(0)2}{\gamma_{46}}], \tag{17}$$

where

$$\overset{(1)}{\gamma_{13}}=\gamma_1[\overset{(1)}{M_y}+\overset{(1)}{\tilde{\tau}_3}L_0],\quad \overset{(0)}{\gamma_{23}}=\gamma_2L_0,\quad \overset{(0)}{\gamma_{46}}=\gamma_2L_0,$$

$$(\gamma_{46}^2)^{(1)}=\gamma_2^2\,2L_0^2\tilde{\tau}_7^{(1)},\quad \overset{(0)}{\gamma_{56}}=0,\quad (\gamma_{56}^2)^{(1)}=0,$$

and all the $\alpha_{ij}$ in (16) and (17) coincide with the $\alpha_{ij}$ from (14) and (15).

It follows therefore that if we assume, despite the absence of direct experimental proof at present, that the phenomenological parameters of anisotropic origin ($\tilde{\Phi}_{7,8}$, $\tilde{\tau}_3$, $\tilde{\tau}_6$, $\tilde{\tau}_7$, $\tilde{\tau}_9$, $\tilde{\tau}_{11}$) are of first rather than zeroth order of smallness, and that $\tilde{\Phi}_{10,7}$ is of second rather than first order, then the equations for the low-frequency and high-frequency AFMR branches in $NiF_2$ at $\mathbf{H}\|[010]$ can be finally written in the form:

$$\begin{aligned}\left(\frac{\omega_1}{\gamma}\right)^2=\frac{\chi_\perp}{\chi_\perp}\Big\{&2H_{A4}H_E+4H_{D\perp}^2-4\alpha H_{D\|}^2+\frac{1}{\alpha}\Big[H_{D\perp}\frac{\Delta\gamma}{\gamma}\\&+2\alpha H_{D\|}-2H_\tau\left(1+\frac{\Delta\gamma}{\gamma}\right)\Big]^2+H\Big[(5H_{D\perp}-4\alpha H_{D\|})\\&+2\left(H_{D\perp}\frac{\Delta\gamma}{\gamma}+2\alpha H_{D\|}-2H_\tau\left(1+\frac{\Delta\gamma}{\gamma}\right)\right)\left(1+\frac{\Delta\gamma}{\gamma}\frac{1}{\alpha}\right)\Big]\\&+H^2\Big[(1-\alpha)+\alpha\left(1+\frac{\Delta\gamma}{\gamma}\frac{1}{\alpha}\right)^2\Big],\end{aligned} \tag{18}$$

$$(\omega_2/\gamma)^2=2H_{A2}H_E+H_{D\perp}^2+4H_{A2}H_{\tau2}+HH_{D\perp}, \tag{19}$$

where

$$\gamma\equiv\tilde{\gamma}_2,\ \Delta\gamma\equiv\tilde{\gamma}_1-\tilde{\gamma}_2,\ H_\tau\equiv{}^1/_2\tilde{\tau}_2H_E,\ H_{\tau2}\equiv\tilde{\tau}_7H_E,\ \alpha\equiv\chi_\|/\chi_\perp,$$
$$H_{D\perp}\equiv-\tilde{\Phi}_7^{(1)}L_0,\ H_{D\|}\equiv-(\tilde{\Phi}_7^{(1)}+{}^1/_2\tilde{\Phi}_8^{(1)}L_0^2)L_0,\ H_{A4}\equiv2\tilde{\Phi}_{10}^{(2)}L_0^3,\ H_{A2}\equiv2\tilde{\Phi}_4^{(1)}L_0.$$

Since we are developing a self-consistent theoretical description based, on the one hand, on actually measured quantities and, on the other, on the accuracies with which they are determined, we must emphasize that these two aspects play different roles. In fact, the lowest-order terms in the expressions for the frequencies, magnetizations, etc., are determined by the values of the frequencies, magnetizations, etc. themselves, while the highest power of the expansion is determined by the present-day experimental accuracy. Once experiments performed with higher accuracy are reported, it will be necessary to add terms of higher powers of $M/L$ in the expressions for the frequencies and magnetizations, to fit the improved experimental accuracy. The numerical values of the phenomenological parameters, obtained from more accurate experiments by new equations that differ from the old ones only in containing terms of higher degree in $M/L$, will not be more precise than the less accurate experiment; the accuracy of the parameters will increase with the experimental accuracy (see Ref. 1).

If we write down the motion matrix $\hat{\gamma}$ in the exchange approximation, i.e., if we set all the $\tau_i$ in (2) equal to zero, then we obtain from (18) and (19) at $H_\tau=H_{\tau2}=0$ the equations previously used to determine the numerical values of the phenomenological parameters of Ref. 7, where it was reported that a value $(\tilde{\gamma}_1-\tilde{\gamma}_2)/\tilde{\gamma}_2=-15\%$ was experimentally observed in $NiF_2$ at $T=4$ K. We note however (see also below) that the real deviation of $\Delta\gamma/\gamma$ is even larger: $\Delta\gamma/\gamma=-45\pm5\%$. The difference between the values of $\Delta\gamma/\gamma$ is due to the fact that when only exchange equations of motion are used the anisotropy enters only via the equilibrium thermodynamic potential. The considerable difference between $\Delta\gamma$ determined without and with allowance for the dynamic anisotropy proves experimentally the need for taking into account the dynamic anisotropy in the matrix $\hat{\gamma}$ when it comes to describing the linear dynamics in substances with unquenched orbital angular momentum.

To describe the behavior of the magnetization and of the AFMR frequencies in the case of arbitrary orientation of $\mathbf{H}$ in the (001) plane, it is convenient to change over the Euler system of coordinates[2]:

$$\begin{aligned}&M_x=S\cos\xi[-\sin\chi(\cos\gamma\cos\theta\cos\varphi-\sin\varphi\sin\gamma)+\cos\chi\sin\theta\cos\varphi],\\&M_y=S\cos\xi[-\sin\chi(\cos\gamma\cos\theta\sin\varphi-\sin\gamma\cos\varphi)+\cos\chi\sin\theta\sin\varphi],\\&M_z=S\cos\xi[\sin\chi\sin\theta\cos\gamma+\cos\chi\cos\theta],\\&L_x=S\sin\xi\sin\theta\cos\varphi,\quad L_y=S\sin\xi\sin\theta\sin\varphi,\\&L_z=S\sin\xi\cos\theta,\quad \mathbf{H}=\{H\cos\varphi_1,\quad H\sin\varphi_1,0\},\end{aligned} \tag{20}$$

where $\theta$ and $\varphi$ are the polar angles of the vector $\mathbf{L}$, $\chi=\arccos 2(\mathbf{L}\cdot\mathbf{M})/S^2\sin 2\xi$, and $\gamma$ is the angle that the projection of $\mathbf{M}$ on a plane perpendicular to $\mathbf{L}$ makes with the line where this plane intersects the plane that passes through $\mathbf{L}$ and $\mathbf{z}$. The equilibrium values of $\xi_0$, $\chi_0$, $\varphi_0$ are determined from the condition that $\Phi$ be a minimum. At $\xi_0\ll1$ and $\chi_0\ll1$ the values of $\xi_0$, $\chi_0$, and $\varphi_0$ are determined from the equations

$$\begin{aligned}&\xi_0(\mathbf{H})=\pi/2+[-H_{D\perp}\cos2\varphi_0+H\sin(\varphi_0-\varphi_1)]/H_E,\\&\chi_0(\mathbf{H})=\pi/2+(\chi_\|/\chi_\perp)[H_{D\|}\sin2\varphi_0+H\cos(\varphi_0-\varphi_1)]/\\&\qquad/[-H_{D\perp}\cos2\varphi_0+H\sin(\varphi_0-\varphi_1)],\\&[-H_{D\perp}\cos2\varphi_0+H\sin(\varphi_0-\varphi_1)][-2H_{D\perp}\sin2\varphi_0-H\cos(\varphi_0-\varphi_1)]\\&+[-2H_{D\|}\cos2\varphi_0+H\sin(\varphi_0-\varphi_1)][H_{D\|}\sin2\varphi_0+H\cos(\varphi_0-\varphi_1)]\chi_\|/\chi_\perp\\&+{}^1/_4 2H_{A4}H_E\sin4\varphi_0=0.\end{aligned} \tag{21}$$

In particular, at $\mathbf{H}\|[110]$ it is easy to obtain from (20) and (21) (see Ref. 1) an expression for the magnetization component $\mathbf{M}_\|$ parallel to $\mathbf{H}$:

$$\begin{aligned}(M_\|/\sigma_{D\perp})=&\frac{\sqrt{2}}{2}+\left(\frac{H}{H_{D\perp}}\right)\frac{1}{2}\Big[(1+\alpha)+\left(\frac{H_{D\perp}-2\alpha H_{D\|}}{H_\Delta}\right)^2\Big]\\&+\left(\frac{H}{H_{D\perp}}\right)^2\frac{3}{2\sqrt{2}}\Big[\frac{H_{D\perp}-2\alpha H_{D\|}}{H_\Delta}\Big]\cdot\\&\cdot\Big[(1-\alpha)-\frac{1}{2}\frac{(H_{D\perp}-2\alpha H_{D\|})(5H_{D\perp}-4\alpha H_{D\|})}{H_\Delta^2}\Big],\\&H_\Delta^2=2H_{A4}H_E-4\alpha H_{D\|}^2+4H_{D\perp}^2.\end{aligned} \tag{22}$$

The experimental plot of $M_\|(H)$ at $\mathbf{H}\|[110]$ in fields up to 65 kOe, shown in Fig. 9 and taken from the paper

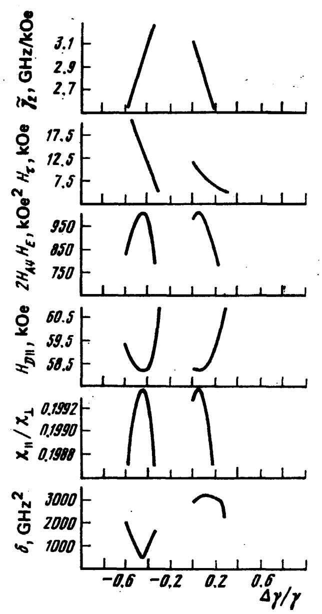


FIG. 2. Dependence of $\delta$, $\chi_\parallel/\chi_\perp$, $H_{D\parallel}$, $2H_{A4}H_E$, $H_\tau$: $\gamma_2$ on $\Delta\gamma/\gamma$.

of Borovik-Romanov *et al.*[14] was reduced by us by least squares using the equation

$$(M_\parallel/\sigma_{D\perp})=A_0+A_1(H/H_{D\perp})+A_2(H/H_{D\perp})^2, \tag{23}$$

where

$\sigma_{D\perp}=$ cgs emu/mol,[14] $\quad H_{D\perp}=27.2$ kOe,[14]
$A_0=0.709\pm0.003,\quad A_1=0.606\pm0.005,\quad A_2=0.078\pm0.002.$

Comparing (11) and (12) with (18), (22) and (23), we obtain five nonlinear equations for six unknown phenomenological parameters ($\chi_\parallel$, $H_{D\parallel}$, $H_{A4}$, $\tilde{\gamma}_2$, $\tilde{\gamma}_1$, $\tilde{\tau}_2$). In place of the sixth equation we use the measured[15] dependence of the frequency of the low-frequency AFMR branch at $\mathbf{H}\parallel[110]$ on the magnetic field. To obtain analytic expressions for the AFMR frequencies at $\mathbf{H}\parallel[110]$, we change in the equations of motion (1) to the coordinate system (20). Then, for an arbitrary direction of $\mathbf{H}$ in the (001) plane, leaving out the intermediate steps given in Ref. 1, we get

$$\left(\frac{\omega_1}{\gamma}\right)^2=\frac{B}{S_0\cos^2\xi_0}\left\{\left[A-2D\frac{\Delta\gamma}{\gamma}+C\left(\frac{\Delta\gamma}{\gamma}\right)^2\right]+\frac{\tilde{\tau}_2}{\cos\xi_0}\left[-2D+2C\frac{\Delta\gamma}{\gamma}+\frac{\tilde{\tau}_2}{\cos\xi_0}\right]\right\}, \tag{24}$$

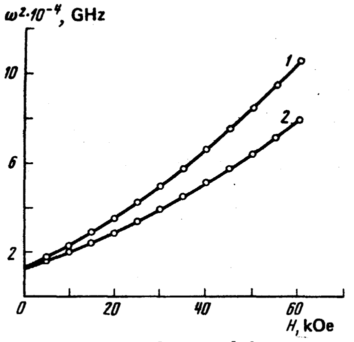


FIG. 3. Dependence of the square of the frequency of the low-frequency branch of AFMR in $NiF_2$ at $T=4.2$ K for the cases $\mathbf{H}\parallel[010]$ curve 1 and $\mathbf{H}\parallel[110]$ (2). The experimental points were taken from Ref. 15 and the curves are plots of Eqs. (18), (21), and (24).

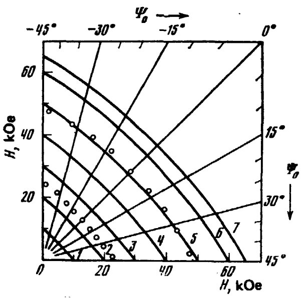


FIG. 4. Dependence of the AFMR resonant field on the angle $\psi_0$ between $\mathbf{H}$ and the [010] axis ($\mathbf{H}\perp[001]$) in $NiF_2$ at $T=4.2$ K and at fixed values of the square of the frequency (cm$^{-2}$): 1) $-22.2$, 2) $-32$, 3) $-43.3$, 4) $-56.7$, 5) $-72.2$, 6) $-89.5$, 7) $-99.1$. The experimental points were taken from 15, and the curves are plots of (21) and (24).

where

$$A=S_0\cos\xi_0[-4H_{D\perp}\cos 2\varphi_0+H\sin(\varphi_0-\varphi_1)]+2H_{A4}\cos 4\varphi_0,$$
$$B=S_0\cos\xi_0(\chi_\perp/\chi_\perp^*)H_E,\quad C=S_0\cos^2\xi_0(\chi_\perp/\chi_\parallel)H_E,$$
$$D=-S_0\cos\xi_0[-2H_{D\parallel}\cos 2\varphi_0+H\sin(\varphi_0-\varphi_1)].$$

The procedure for determining the numerical values of the phenomenological parameters of the theory from the experimental data, which is described in detail in Ref. 1, consists of finding, at fixed values of $-1<\Delta\gamma/\gamma<1$, values of $H_{D\parallel}$, $\chi_\parallel$, $H_{A4}$, $\tilde{\gamma}_1$, $\tilde{\gamma}_2$, $\tilde{\tau}_2$, that satisfy expressions (11), (12), (18), (22), and (23), and at the same time minimize the functional

$$\delta=\left[\sum_i(\omega_i^2-\omega_{ie}^2)^2/N\right]^{1/2},$$

which characterizes the deviation of the theoretical function (24) ($\mathbf{H}\parallel[110]$) from the experimental one.[15] Figure 2 shows plots of $\delta$, $\chi_\parallel/\chi_\perp$, $H_{D\parallel}$, $\tilde{\gamma}_2$, $H_\tau$ against $\Delta\gamma/\gamma$. It is seen that at $\Delta\gamma/\gamma=-0.45$ the mean squared deviation $\delta$ has a clearly pronounced minimum, $\chi_\parallel/\chi_\perp=0.199$, $H_{D\parallel}=58.2$ kOe, $2H_{A4}H_E=1010$ kOe$^2$, $H_\tau=14.1$ kOe, and $\tilde{\gamma}_2=2.98$ GHz/kOe. The numerical values of the phenomenological parameters make it possible to plot, for any orientation of the magnetic field in the (001) plane, the AFMR frequencies (Figs. 3 and 4), the magnetizations (Figs. 5 and 6), $\xi_0$ and $\chi_0$ (Fig. 7), and $\varphi_0$ (Fig. 8). The experimental and calculated curves agree within the limits of the accuracy of the measurements and of the calculations.

Comparing the theoretical (19) and the experimental[16] dependences of the high-frequency AFMR branches for $\mathbf{H}\parallel[010]$, we obtain $H_{D\perp}=28\pm9$ kOe and $H_{A2}=40\pm2$

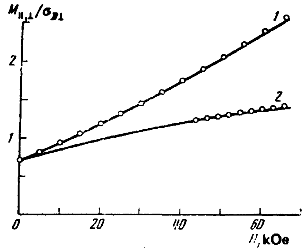


FIG. 5. Plots of the magnetizations parallel (curve 1) and perpendicular (2) to the magnetic field $\mathbf{H}\parallel[110]$ against $\mathbf{H}$ in $NiF_2$ at $T=4.2$ K. The experimental points were taken from Ref. 14 and the curves are plots of Eqs. (20) and (21).

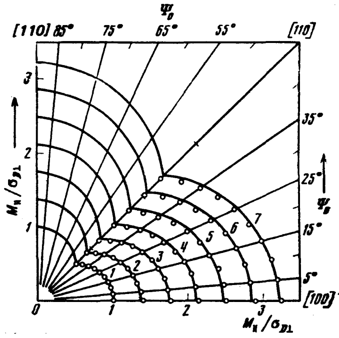

FIG. 6. Plots of the magnetization parallel ($M_{\parallel}$) to the magnetic field $\mathbf{H}\perp[001]$ against the angle $\Psi_0$ between $\mathbf{H}$ and [010] in $NiF_2$ at $T=4.2$ K and at fixed values of the magnetic field (in kOe): 1) – 0, 2) – 10, 3) – 20, 4) – 30, 5) – 40, 6) – 50, 7) – 60. The experimental points were taken from Ref. 14, at the curves are plots of (20) and (21).

kOe.

Thus, using an actual antiferromagnet as an example, we have shown that the maximum number of phenomenological parameters in a model-independent theory is determined by the symmetry and is limited in principle by the experimental accuracy. By comparing the theoretical equations obtained in the present paper with the four experimental functions $\mathbf{M}=M(\mathbf{H}\|[110])$, $\omega_1=\omega_1(\mathbf{H}\|[100])$, $\omega_1=\omega_1(\mathbf{H}\|[110])$, $\omega_2=\omega_2(\mathbf{H}\|[100])$ we determined the numerical values of seven phenomenological parameters: $\tilde{\gamma}_1, \tilde{\gamma}_2, \tilde{\tau}_2, \tilde{\Phi}_8, \tilde{\Phi}_3, \tilde{\Phi}_{10}, \tilde{\Phi}_4$. Together with the previously known parameters $\tilde{\Phi}_2, \tilde{\Phi}_5, \tilde{\Phi}_7$ the parameters $\tilde{\Phi}_2, \tilde{\Phi}_3, \tilde{\Phi}_4, \tilde{\Phi}_5, \tilde{\Phi}_7, \tilde{\Phi}_8, \tilde{\Phi}_{10}, \tilde{\gamma}_1, \tilde{\gamma}_2, \tilde{\tau}_2$ constitute a complete set of phenomenological parameters that make it possible to construct the potential and the equations of motion. The constructed potential and equations of motion contain finite numbers of terms and describe adequately the static and the dynamic properties of $NiF_2$ at $T=4.2$ K. With the aid of the constructed equations of motion and the potential, we calculated the values of the magnetization components and the AFMR frequencies for an arbitrary orientation of the magnetic field in the (001) plane. In the entire interval of magnetic fields ($H\leqslant 65$ kOe) for which reliable magnetization measurements are available at present, the experimental and calculated plots of the magnetization and of the AFMR frequencies agree within the limits of the accuracy of the measurement and the calculations.

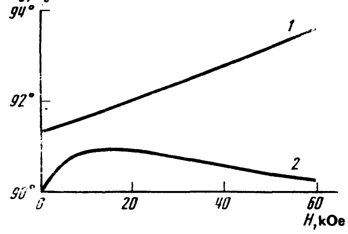

FIG. 7. Plots of the angles $\xi_0$ (curve 1) and $\chi_0$ (curve 2), calculated from Eqs. (21), against the magnetic field $\mathbf{H}|$ [110] in $NiF_2$ at $T=4.2$ K.

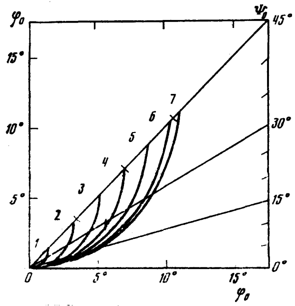

FIG. 8. Plots, calculated from (21), of the angle $\xi_0$ against the angle $\chi_0$ between $\mathbf{H}$ and the [010] axis in $NiF_2$ at $T=4.2$ K and at fixed values of the magnetic field (kOe): 1) – 10, 2) – 20, 3) – 30, 4) – 40, 5) – 50, 6) – 60, 7) – 65.

It is of interest to compare the descriptions of the low-frequency AFMR branch in $NiF_2$ at $T=4.2$ K obtained by the non-equilibrium-thermodynamics and Lagrangian approaches. To this end, we rewrite Eq. (18) in the form $\omega_1^2=\Omega_1^2+\Omega_2^2$, where

$$\Omega_1^2=\gamma_2^2(\chi_\perp/\chi_\perp^*)[H_{D\perp}(\Delta\gamma/\gamma)+2\alpha H_{D\parallel}-2H_\tau(1+\Delta\gamma/\gamma)+\alpha H(1+\Delta\gamma/(\gamma\alpha))]^2(1/\alpha),$$
$$\Omega_2^2=\gamma_2^2(\chi_\perp/\chi_\perp^*)[2H_{A1}H_E+4H_{D\perp}^2-4\alpha H_{D\parallel}^2+(5H_{D\perp}-4\alpha H_{D\parallel})H+H^2(1-\alpha)]. \quad (25)$$

The expression for $\Omega_2$ coincides with the dependence of the frequency on the magnetic field, obtained in Ref. 11 by the Lagrangian approach under the assumption $\rho_1\sim\rho_2$ (see Ref. 1) at $\tilde{\gamma}_2=1/\rho_2 L_0^2$. In the Lagrangian approach at an arbitrary value of the magnetic field we have $\Omega_1=0$, which leads to the existence of a connection between the static ($\chi_\parallel, H_E, H_{D\perp}, H_{D\parallel}$) and dynamic ($H_\tau, \tilde{\gamma}_1, \tilde{\gamma}_2$) phenomenological parameters at $T=0$ K. In fact, equating separately to zero the coefficients of $H$ in the zeroth and first powers in Eq. (25) for $\Omega_1$, we obtain

$$H_{D\perp}(\Delta\gamma/\gamma)+2(\chi_\parallel/\chi_\perp)H_{D\parallel}-2H_\tau(1+\Delta\gamma/\gamma)=0,\quad 1+\frac{\Delta\gamma}{\gamma}\frac{\chi_\perp}{\chi_\parallel}=0,$$

from which it follows that

$$\frac{\Delta\gamma}{\gamma}=-\frac{\chi_\parallel}{\chi_\perp},\quad H_\tau=\frac{2H_{D\parallel}-H_{D\perp}}{2(1-\chi_\parallel/\chi_\perp)}\frac{\chi_\parallel}{\chi_\perp}.$$

Using the presently obtained numerical values of the phenomenological parameters, we can directly calculate $\Omega_1$ and $\Omega_2$ at $T=4.2$ K. Figure 9 shows the dependence of the ratio $\Omega_1^2/\Omega_2^2$ on the magnetic field ($H\leqslant 65$ kOe).

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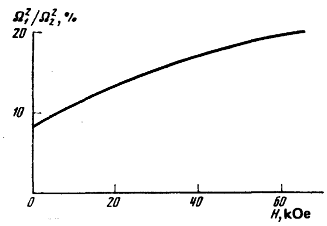

FIG. 9. Ratio of the squared frequencies $\Omega_1^2$ and $\Omega_2^2$, calculated from (25), against the magnetic field $\mathbf{H}\|[010]$ at $T=4.2$ K.

constant attention, support, and a discussion of the paper.


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