Magnetodipole broadening of a resonance line in a superparamagnet

R. S. Gekht and V. I. Ignatchenko

L. V. Kirenskiĭ Physics Institute, Siberian Division, USSR Academy of Sciences
(Submitted 26 May 1978; resubmitted 1 September 1978)
Zh. Eksp. Teor. Fiz. 76, 164–174 (January 1979)

The influence of magnetodipole interaction on the shape and width of a resonance line, and also on the average magnetization curve of a superparamagnet (a system of minute single-domain ferromagnetic particles) is considered. Using a classical analog of the van Vleck quantum method of moments, expressions are obtained for the first four moments of the absorption line of a superparamagnet in which the particles are randomly arranged; these expressions are valid in a wide temperature interval. It is established that in a region where the ferromagnetic-particle concentration is very low, $c \sim 10^{-3}$, the lines have a quasi-Lorentz shape and with increasing concentration (at $c \sim 0.1$), a Gaussian shape is approached; for both shapes, expressions are obtained for the width of the resonance line as a function of the temperature, volume, and concentration of the particles. The results for the magnetodipole linewidth are compared with previously derived expressions for the linewidth due to the internal relaxation of the magnetization in the particles. It is shown that the average magnetization curve of a superparamagnet, calculated with allowance for the magnetodipole interaction, lies below the Langevin curve in the entire temperature interval.

PACS numbers: 75.20. – g, 75.60.Jp, 75.60.Ej

INTRODUCTION

The width of the magnetic-resonance line in an ideal superparamagnet, i.e., in an ensemble of noninteracting ferromagnetic single-domain particles, was investigated by Raĭkher and Shliomis[1] (resonance in the anisotropy field in the absence of an external stationary magnetic field) as well as in Refs. 2 and 3 (resonance in an external stationary magnetic field). In such a system, the finite magnetic-resonance line width is due to internal mechanisms in each paramagnetic particle (spin–lattice interaction, etc.), which can be described phenomenologically at $T = 0$ by the damping term in the Landau–Lifshitz equation.[4] With increasing intensity of the thermal fluctuations and with decreasing particle dimensions, broadening of the absorption lines takes place; the line shape does not change in this case and is always Lorentzian.

The purpose of the present work is to investigate the influence of the magnetodipole interaction between particles on the shape and width of the radio-frequency field absorption line; this interaction increases with increasing concentration of the ferromagnetic particles in the nonmagnetic matrix, and neglect of this interaction is no longer valid at certain concentrations.

A similar problem for quantum paramagnetic systems was solved by van Vleck.[5] In view of the difficulties that arise when attempts are made to obtain an analytic expression for the absorption line, van Vleck proposed a method that consists of calculating the first few moments of the absorption line and reconstructing approximately, from these moments, the shape and width of the resonance line; this procedure was subsequently named the "van Vleck method of moments." The main idea of this method will be used in the present paper. However, in the study of nuclear and electron resonances in paramagnets, the high-temperature approximation for the density matrix is always sufficient, since the magnetic energy of the nuclei and of the electrons in an external magnetic field is much lower than the thermal energy. On the other hand, a distinguishing feature of ferromagnetic particles is that they have a considerable magnetic moment, and the Zeeman energy can be comparable with the thermal energy.

To describe this situation, we develop in this paper a classical analog of van Vleck's quantum method of moments, valid in a wide range of temperatures. In the first section we write down an equation for the probability function W of the orientation of the magnetic moments. Since the magnetic moments of the ferromagnetic particles are large, they can regarded as classical spins described by the Liouville equation. We assume that the superparamagnet is in a sufficiently strong magnetic field and therefore the magnetodipole interaction between the ferromagnetic particles is weak compared with the Zeeman energy. In this approximation, we obtain general expressions for the first four moments of the absorption line as functions of the temperature, particle dimension, and particle volume concentration. We obtain also the average value of the projection of the magnetic moment of the superparamagnetic system on the direction of the stationary magnetic field. In the second section we assess these expressions numerically and analytically for different limiting cases. We consider also the joint action of the internal and magnetodipole relaxations.

1. METHOD OF MOMENTS FOR A SUPERPARAMAGNET

Assume a system of N ferromagnetic particles in a single-domain state. The dynamic behavior of the magnetization of each ferromagnetic particle is then described by the Landau–Lifshitz equation,[4] which in

 0038-5646/79/010084-06$02.40

a spherical coordinate system can be easily reduced to the Hamiltonian form[6]:

$$\frac{d(\mathcal{M}^i\cos\theta_i)}{dt}=\frac{\partial\mathcal{H}}{\partial(\varphi_i/\gamma_i)}\qquad\frac{d(\varphi_i/\gamma_i)}{dt}=-\frac{\partial\mathcal{H}}{\partial(\mathcal{M}^i\cos\theta_i)}. \tag{1.1}$$

Here $\mathcal{H}$ is the Hamiltonian of the system expressed in terms of the generalized coordinates $q_i=\mathcal{M}^i\cos\theta_i$ and momenta $p_i=\varphi_i/\gamma_i$; $\mathcal{M}^i$ is the magnetic moment of the i-th ferromagnetic particles. θ_i and φ_i are the polar and azimuthal angles of the magnetic moment, and γ_i is the gyromagnetic ratio.

The properties of a system of N interacting ferromagnetic particles are described by the kinetic Liouville equation

$$\frac{\partial W}{\partial t}=\{\mathcal{H},W\}=\sum_{i=1}^{N}\left(\frac{\partial\mathcal{H}}{\partial q_i}\frac{\partial W}{\partial p_i}-\frac{\partial\mathcal{H}}{\partial p_i}\frac{\partial W}{\partial q_i}\right). \tag{1.2}$$

Using the expression for the Poisson brackets (1.2), we easily obtain in the classical case permutation relations for the projections of the magnetic moment $\mathcal{M}^i_\alpha(\alpha=x,y,z)$, which are the analog of the quantum commutation relations:

$$\{\mathcal{M}_\alpha^i,\mathcal{M}_\beta^j\}=-\gamma_i\mathcal{M}_\tau^i\delta_{ij}\varepsilon_{\alpha\beta\tau}. \tag{1.3}$$

Here δ_{ij} is the Kronecker symbol, $\varepsilon_{\alpha\beta\gamma}$ is a unit antisymmetrical tensor.

Below, however, we shall need also more general permutation relations

$$\{\mathcal{M}_\alpha^i,f(\mathcal{M}_\beta^j)\}=-\gamma_i\mathcal{M}_\tau^i\delta_{ij}\varepsilon_{\alpha\beta\tau}\frac{\partial f(x)}{\partial x}\Big|_{x=\mathcal{M}_\beta^j}, \tag{1.4}$$

which are obtained by expanding the arbitrary function $f(\mathcal{M}_\beta^j)$ in powers of the argument $\mathcal{M}_\beta^j$ using the properties of the Poisson brackets.

The Liouville equation (1.2) is satisfied by the equilibrium Gibbs function

$$W_0=Z^{-1}\exp(-\mathcal{H}/kT), \tag{1.5}$$

where

$$Z=\int\exp(-\mathcal{H}/kT)\,d\Gamma,\qquad d\Gamma=dq_1dp_1dq_2dp_2\ldots dq_Ndp_N.$$

We consider now an ensemble of N identical ferromagnetic particles of spherical form with volume V, magnetization M, and magnetic isotropy. Let the ferromagnetic particles with magnetic moment $\mathcal{M}=MV$ be situated in a solid nonmagnetic medium, in which they are randomly distributed; a stationary magnetic field H_0 is applied along the z axis. The Hamiltonian of the system, with allowance for the magnetodipole interaction $\mathcal{H}_d$, is of the form

$$\mathcal{H}=\mathcal{H}_0+\mathcal{H}_d=-H_0\sum_{j=1}^{N}\mathcal{M}_z^j$$
$$+\frac{1}{2}\sum_{j\neq k,\alpha,\beta}\frac{1}{r_{jk}^3}\left[\mathcal{M}_\alpha^j\mathcal{M}_\alpha^k-3\frac{(\mathcal{M}_\alpha^j r_{jk}^\alpha)(\mathcal{M}_\beta^k r_{jk}^\beta)}{r_{jk}^2}\right]. \tag{1.6}$$

We assume that the energy of the interaction of the magnetic moments with the external stationary field H_0 greatly exceeds the energy of the dipole–dipole interaction, i.e., $\mathcal{H}_d$ is a small perturbation.

Calculating the average value of the function

$$m_z=\sum_{j=1}^{N}\mathcal{M}_z^j/N\mathcal{M}$$

over the equilibrium Gibbs function (1.5) and averaging the obtained expression over the particles that are randomly distributed in space, we have

$$\langle m_z\rangle=\langle m_z\rangle_0-\frac{\pi^2}{45}c\left(\frac{M^2V}{kT}\right)^2[8\langle m_z\rangle_0^3(\langle m_z^2\rangle_0-\langle m_z\rangle_0^2)$$
$$+(14-16.5\langle m_z^2\rangle_0)(\langle m_z^2\rangle_0-\langle m_z\rangle_0\langle m_z^2\rangle_0)], \tag{1.7}$$

where $c=NV/V_c$ is the volume concentration of the ferromagnetic particles; V_v is the volume of the superparamagnetic system, that includes the particles in the matrix. The symbol $\langle\ldots\rangle$ will denote averaging over the equilibrium function with total Hamiltonian $\mathcal{H}_0+\mathcal{H}_d$, and the symbol $\langle\ldots\rangle_0$ denotes averaging over the equilibrium function with Zeeman Hamiltonian $\mathcal{H}_0$ without allowance for the dipole–dipole interaction; the last mean values are defined by the expressions

$$\langle m_z\rangle_0=\operatorname{cth}\sigma-\sigma^{-1}\equiv L(\sigma),\qquad\langle m_z^2\rangle_0=1-\frac{2}{\sigma}\operatorname{cth}\sigma+\frac{2}{\sigma^2},$$
$$\langle m_z^3\rangle_0=\operatorname{cth}\sigma-\frac{3}{\sigma}+\frac{6}{\sigma^2}\operatorname{cth}\sigma-\frac{6}{\sigma^3}, \tag{1.8}$$

where $L(\sigma)$ is the Langevin function with argument $\sigma=H_0MV/kT$.

At high temperatures, the deviation of the average magnetization from the Langevin function is

$$\langle m_z\rangle-L(\sigma)\approx-\frac{\pi^2}{60}c\left(\frac{M^2V}{kT}\right)^2\frac{H_0MV}{kT}, \tag{1.9}$$

and at low temperatures

$$\langle m_z\rangle-L(\sigma)\approx-\pi^2c\left(0.42+0.09\frac{kT}{H_0MV}\right)\left(\frac{M}{H_0}\right)^2. \tag{1.10}$$

A plot of the function (1.7) in the entire temperature region is shown in Fig. 1 by the dashed curve for the value $M\sqrt{c}/H_0=0.1$. The solid line in the same figure shows the Langevin function $L(\sigma)$. Because of the disorder caused by the dipole–dipole interaction, the dashed curve lies lower than the Langevin curve in the entire temperature region.

We consider now the magnetic resonance in such a system; let a weak radio frequency field $h\cos\omega t$ be applied along the x axis. Then the Hamilton of the interaction with the external field is

$$\mathcal{H}_1=-\mathcal{M}_xh\cos\omega t,\qquad\mathcal{M}_x=\sum_{j=1}^{N}\mathcal{M}_x^j. \tag{1.11}$$

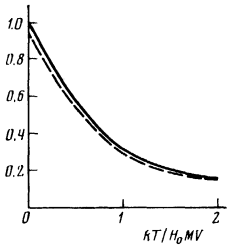

FIG. 1. Temperature dependence of the average magnetization of a superparamagnetic system $\langle m_z\rangle$. Solid—without allowance for the magetodipole interaction (Langevin function), dashed—with allowance for the magetodipole interaction (expression (1.7)).

The imaginary part of the linear susceptibility is given by the Kubo formula[7]

$$\chi''(\omega)=\int_0^\infty G(t)\sin\omega t dt, \tag{1.12}$$

where the relaxation function $G(t)$ is given by

$$G(t)=\langle\{\exp[\hat{L}(\mathcal{H})t]\mathcal{M}_x,\ \mathcal{M}_x\}\rangle. \tag{1.13}$$

Here $\hat{L}(\mathcal{H})$ is the Liouville operator

$$\hat{L}(\mathcal{H})=\sum_{i=1}^{N}\left(\frac{\partial\mathcal{H}}{\partial q_i}\frac{\partial}{\partial p_i}-\frac{\partial\mathcal{H}}{\partial p_i}\frac{\partial}{\partial q_i}\right). \tag{1.14}$$

Just as in Ref. 5, we assume that the energy of the dipole–dipole interaction is small compared with the Zeeman energy, i.e.,

$$H_0\gg MV/\bar{r}^3, \tag{1.15}$$

where $\bar{r}$ is the average distance between the ferromagnetic particles. Satisfaction of this condition makes it possible[5] to simplify the Hamiltonian (1.6) and retain in $\mathcal{H}_d$ only the principal part $\mathcal{H}_d'$ which makes the main contribution to the line broadening in the vicinity of the resonant frequency $\omega_0=\gamma H_0$. To separate $\mathcal{H}_d'$ in our case it is necessary to satisfy the condition $\{\mathcal{H}_0,\mathcal{H}_d'\}=0$, which corresponds to averaging over the fast variable in the Landau–Lifshitz equations (1.1). As a result we have, instead of the initial Hamiltonian $\mathcal{H}_d$ of the dipole–dipole interaction, just as in Ref. 5,

$$\mathcal{H}_d'=\frac{1}{6}\sum_{j\neq k}b_{jk}(3\mathcal{M}_z^j\mathcal{M}_z^k-\mathcal{M}_\alpha^j\mathcal{M}_\alpha^k), \tag{1.16}$$

where

$$b_{jk}=\frac{3}{2}\frac{1-3\cos^2\vartheta_{jk}}{r_{jk}^3} \tag{1.17}$$

and ϑ_{jk} is the polar angle of the vector $\mathbf{r}_{jk}$. Using the permutation relations (1.3), we easily see that $\{\mathcal{H}_0,\mathcal{H}_d'\}=0$.

According to (1.14), the expression for the Liouville operator of the Zeeman Hamiltonian $\mathcal{H}_0$ is

$$\hat{L}(\mathcal{H}_0)=-\omega_0\sum_{j=1}^{N}\frac{\partial}{\partial\varphi_j}. \tag{1.18}$$

Expanding $\exp[\hat{L}(\mathcal{H}_0)t]$ in a series, differentiating term by term with respect to $\partial\varphi_j$, and combining the obtained series into functions of $\cos\omega_0 t$ and $\sin\omega_0 t$, we obtain

$$\exp[\hat{L}(\mathcal{H}_0)t]\mathcal{M}_x=\mathcal{M}_x\cos\omega_0 t+\mathcal{M}_y\sin\omega_0 t. \tag{1.19}$$

Substituting in (1.13) $\mathcal{H}=\mathcal{H}_0+\mathcal{H}_d'$ and using (1.19), we obtain from (1.12) the final expression for the susceptibility

$$\chi''(\omega)=\int_0^\infty(G_1(t)\cos\omega_0 t+G_2(t)\sin\omega_0 t)\sin\omega t dt. \tag{1.20}$$

The functions $G_1(t)$ and $G_2(t)$ are the abbreviated classical relaxation functions

$$\begin{aligned}G_1(t)&=\langle\{\exp[\hat{L}(\mathcal{H}_d')t]\mathcal{M}_x,\ \mathcal{M}_x\}\rangle,\\ G_2(t)&=\langle\{\exp[\hat{L}(\mathcal{H}_d')t]\mathcal{M}_y,\ \mathcal{M}_x\}\rangle.\end{aligned} \tag{1.21}$$

The moments of the susceptibility curve $\chi''(\omega_0+\Omega)$ near the resonant frequency ω_0 are determined by the following expressions[8]: for the odd moments

$$\langle\Omega^{2n+1}\rangle=(-1)^n\frac{1}{G_2(0)}\frac{d^{2n+1}G_1(t)}{dt^{2n+1}}\Big|_{t=0},\quad \Omega=\omega-\omega_0, \tag{1.22a}$$

and for the even moments

$$\langle\Omega^{2n}\rangle=(-1)^n\frac{1}{G_2(0)}\frac{d^{2n}G_2(t)}{dt^{2n}}\Big|_{t=0}. \tag{1.22b}$$

Substituting (1.21) in (1.22a) and (1.22b) we obtain expressions for the first and second moments

$$\langle\Omega\rangle=\frac{\langle\{\{\mathcal{H}_d',\mathcal{M}_x\},\mathcal{M}_x\}\rangle}{\langle\{\mathcal{M}_y,\mathcal{M}_x\}\rangle}=\frac{H_0}{kT}\frac{\langle\mathcal{M}_y\{\mathcal{H}_d',\mathcal{M}_x\}\rangle}{\langle\mathcal{M}_x\rangle}, \tag{1.23a}$$

$$\langle\Omega^2\rangle=-\frac{\langle\{\{\mathcal{H}_d',\{\mathcal{H}_d',\mathcal{M}_y\}\},\mathcal{M}_x\}\rangle}{\langle\{\mathcal{M}_y,\mathcal{M}_x\}\rangle}=\frac{H_0}{kT}\frac{\langle\{\mathcal{H}_d',\mathcal{M}_y\}^2\rangle}{\langle\mathcal{M}_x\rangle}. \tag{1.23b}$$

We have used here the permutation properties under the mean-value symbol, i.e.,

$$\int_\Gamma A\cdot\{B,C\}d\Gamma=\int_\Gamma\{A,B\}\cdot Cd\Gamma,$$

where A, B, and C are arbitrary dynamic variables, as well as the expressions

$$\{\mathcal{M}_x,W_0\}=\frac{\omega_0}{kT}\mathcal{M}_yW_0,\quad \{\mathcal{M}_y,\mathcal{M}_x\}=\gamma\mathcal{M}_z,\quad \{W_0,\mathcal{H}_d'\}=0,$$

which are obtained from the permutation relations for the Poisson brackets (1.3) and (1.4); the last expression follows also from the condition that the Poisson brackets of $\mathcal{H}_0$ and $\mathcal{H}_d'$ vanish.

The most substantial difference between the expression for the second moment (1.23) (as well as the expression for the fourth moment, which will be obtained later on) from the corresponding van Vleck expression[5] is that now we carry out classical averaging over the equilibrium function $W_0(t)$ at an arbitrary temperature T, instead of the quantum averaging in the high-temperature limit. As a result of the smallness of the energy of the dipole–dipole interaction it suffices here, just as in the quantum case, to average over the equilibrium function W_0, which contains only the Hamiltonian $\mathcal{H}_0$ of the Zeeman energy. Calculating the Poisson brackets

$$\{\mathcal{H}_d',\mathcal{M}_x\}=-\gamma\sum_{j\neq k}b_{jk}\mathcal{M}_z^j\mathcal{M}_y^k,\quad \{\mathcal{H}_d',\mathcal{M}_y\}=\gamma\sum_{j\neq k}b_{jk}\mathcal{M}_z^j\mathcal{M}_x^k \tag{1.24}$$

and substituting them in expression (1.23a) and (1.23b) we obtain after averaging the Gibbs function W_0

$$\langle\Omega\rangle=-\gamma MVLN^{-1}\sum_{j\neq k}b_{jk}, \tag{1.25a}$$

$$\langle\Omega^2\rangle=(\gamma MV)^2N^{-1}\left(\langle m_z^2\rangle_0\sum_{j\neq k}b_{jk}^2+\langle m_z\rangle_0^2\sum_{j,k,p\neq}b_{jk}b_{kp}\right), \tag{1.25b}$$

where $j,k,p\neq$ means that j, k, p are not equal to each other, and the mean values $\langle\ldots\rangle_0$ are defined by formulas (1.8). In the derivation of (1.25), account was taken of the fact that the ferromagnetic particles are identical and therefore the corresponding average projections of the moments with different indices are equal to one another and can be taken outside the summation sign:

$$\sum_{\substack{j\neq k\\ p\neq q}}b_{jk}b_{pq}\langle\mathcal{M}_z^j\mathcal{M}_x^k\mathcal{M}_z^p\mathcal{M}_x^q\rangle=\mathcal{M}^4\langle m_z^2\rangle_0\langle m_x^2\rangle_0\sum_{j\neq k}b_{jk}^2+\mathcal{M}^4\langle m_z\rangle_0^2\langle m_x^2\rangle_0\sum_{j,k,p\neq}b_{jk}b_{jp}. \tag{1.26}$$

We have used also the equation

$$\langle m_x^2\rangle_0=(1-\langle m_z^2\rangle_0)/2=\langle m_z\rangle_0/\sigma. \tag{1.27}$$

Similarly, for the third and fourth moment we obtain the expressions

$$\langle\Omega^3\rangle=-\frac{H_0}{kT}\frac{\langle\{\{\mathcal{H}_d',\mathcal{M}_y\}\{\mathcal{H}_d',\{\mathcal{H}_d',\mathcal{M}_z\}\}\rangle}{\langle\mathcal{M}_z\rangle}, \tag{1.28a}$$

$$\langle\Omega^4\rangle=\frac{H_0}{kT}\frac{\langle\{\{\mathcal{H}_d',\{\mathcal{H}_d',\mathcal{M}_y\}\}^2\rangle}{\langle\mathcal{M}_z\rangle}. \tag{1.28b}$$

Substituting (1.24) and using the permutation relations (1.3), we have

$$\begin{aligned}\{\mathcal{H}_d',\{\mathcal{H}_d',\mathcal{M}_z\}\}&=-\frac{1}{3}\gamma^2\sum_{\substack{j\neq k,\\ j\neq p}}b_{jk}b_{jp}(2\mathcal{M}_z^j\mathcal{M}_z^k\mathcal{M}_z^p\\&+\mathcal{M}_z^j\mathcal{M}_x^k\mathcal{M}_x^p+\mathcal{M}_x^j\mathcal{M}_y^k\mathcal{M}_y^p-\mathcal{M}_y^j\mathcal{M}_x^k\mathcal{M}_y^p),\\ \{\mathcal{H}_d',\{\mathcal{H}_d',\mathcal{M}_y\}\}&=-\frac{1}{3}\gamma^2\sum_{\substack{j\neq k,\\ j\neq p}}b_{jk}b_{jp}(2\mathcal{M}_y^j\mathcal{M}_z^k\mathcal{M}_z^p\\&+\mathcal{M}_z^j\mathcal{M}_y^k\mathcal{M}_z^p+\mathcal{M}_y^j\mathcal{M}_z^k\mathcal{M}_z^p-\mathcal{M}_z^j\mathcal{M}_y^k\mathcal{M}_z^p).\end{aligned} \tag{1.29}$$

Substituting these expression in (1.28a) and (1.28b), and calculating the average over the equilibrium function W_0, we obtain the general expressions for the third and fourth moments:

$$\begin{aligned}\langle\Omega^3\rangle=&-\frac{1}{3}(\gamma MV)^3N^{-1}\Big[(3\langle m_z^3\rangle_0+\langle m_z^2m_x\rangle_0)\sum_{j\neq k}b_{jk}^3\\&+L(9\langle m_z^3\rangle_0+\langle m_x^3\rangle_0)\sum_{j,k,p\neq}b_{jk}^2b_{jp}+L^2\sum_{j,k,p,s\neq}b_{jk}b_{jp}b_{js}\Big],\end{aligned} \tag{1.30a}$$

$$\begin{aligned}\langle\Omega^4\rangle=&\frac{1}{9}(\gamma MV)^4N^{-1}\Big[\Big(4\langle m_z^4\rangle_0+\frac{\langle m_z^2\rangle_0}{\langle m_x^2\rangle_0}\langle m_x^3m_z^3\rangle_0+\langle m_z^2\rangle_0\\&+3\langle m_x^3m_z^3\rangle_0\Big)\sum_{j\neq k}b_{jk}^4+(13\langle m_z^2\rangle_0^2+4\langle m_x^2\rangle_0^2+4\langle m_z^2\rangle_0\langle m_x^2\rangle_0)\sum_{j,k,p\neq}b_{jk}^2b_{jp}^2\\&+(9\langle m_z^2\rangle_0^2-3\langle m_x^2\rangle_0^2+4L\langle m_z^3\rangle_0)\sum_{j,k,p\neq}b_{jk}^2b_{jp}b_{kp}+4\langle m_x^2\rangle_0L^2\sum_{j,k,p,r\neq}b_{jk}^2b_{jp}b_{jr}\\&+2\langle m_x^2\rangle_0L^2\sum_{j,k,p,r\neq}b_{jk}^2b_{jp}b_{pr}+20L\langle m_z^3\rangle_0\sum_{j,k,p\neq}b_{jk}^2b_{jp}\\&+L^4\Big(4\sum_{j,k,p,r,t\neq}b_{jk}b_{jp}b_{jr}b_{jt}+\sum_{j,k,p,r,t\neq}b_{jk}b_{jp}b_{rk}b_{rt}\\&+4\sum_{j,k,p,r,t\neq}b_{jk}b_{jp}b_{jr}b_{rt}\Big)\Big].\end{aligned} \tag{1.30b}$$

2. SHAPE AND WIDTH OF RESONANCE ABSORPTION LINE

In the limiting case of high temperatures ($\sigma=H_0MV/kT\ll1$), substituting in (1.25) and (1.30) the high-temperature mean values

$$\begin{aligned}&\langle m_z^2\rangle_0=\langle m_x^2\rangle_0={}^1/_2,\quad \langle m_x^2m_z^2\rangle_0={}^1/_{15},\\&\langle m_z\rangle_0=\langle m_z^3\rangle_0=\langle m_z^2m_x\rangle_0=0,\end{aligned} \tag{2.1}$$

we obtain for the second and fourth moments the expressions

$$\begin{aligned}\langle\Omega^2\rangle=&-\frac{1}{3}(\gamma MV)^2\sum_k b_{jk}^2,\\ \langle\Omega^4\rangle=&\frac{1}{9}(\gamma MV)^4\Big(\frac{7}{5}\sum_k b_{jk}^4+\frac{7}{3}\sum_{k,p\neq}b_{jk}^2b_{jp}^2+\frac{2}{3}\sum_{k,p\neq}b_{jk}^2b_{jp}b_{kp}\Big).\end{aligned} \tag{2.2}$$

These expressions correspond to the classical limit of the results of van Vleck[5] on the dipole broadening of lines in crystals, provided the sbustitution $MV=g\mu_BS$ is made, where g is the Landé factor, μ_B is the electron or nuclear Bohr magneton, and S is the spin quantum number. The odd numbers in the high-temperature approximation are equal to zero.

We turn now to the general expressions for the moments (1.25a), (1.25b) and (1.30a), (1.30b). Let all the particles in the superparamagnet be distributed in random fashion. Then, after averaging expressions (1.25a), (1.25b) and (1.30a), (1.30b) over all the possible configurations of the ferromagnetic particles, we find that in the general case all the moments differ from zero. The expression for the first moment is the shift of the resonance line relative to the frequency $\omega_0=\gamma H_0$, and depends both on the temperature and on the shape of the sample. If the superparamagnet is an ellipsoid of revolution about the z axis, then the temperature dependence of this shift is determined in the classical case by a Langevin function and is given by the following expression:

$$\langle\Omega\rangle=6\pi\gamma McL({}^1/_3-n_z),$$

where n_z is the demagnetizing factor along the z axis. This expression corresponds to the classical limit of the expression for the shift of the resonance line in paramagnetic crystals with cubic structure, obtained in the quantum analysis by McMillan and Opechowski.[9]

With decreasing temperature, the third central moment, $\langle(\Omega-\langle\Omega\rangle)^3\rangle$, contributes to the asymmetry of the resonance-line shape. As seen from (1.30a), owing to the presence of the term with the factor $\sum_{j\neq k}b_{jk}^3$, the shape of resonance line becomes asymmetrical with decreasing temperature even if the superparamagnet sample is spherical.

We shall assume below that the superparamagnet is spherical and neglect the asymmetry of the resonance line, inasmuch as in accord with (1.20) the abbreviated relaxation function $G_1(t)$ for the odd moments enters in a term with a rapidly oscillating factor $\cos\omega_0t\sin\omega t$ and makes a smaller contribution than the second term in the expression for $\chi''(\omega)$. After averaging (1.25b) and (1.30b) over all the possible configuration of the ferromagnetic particles, we get

$$\langle\Omega^2\rangle=(\gamma MV)^2\langle m_z^2\rangle_0\sum_k\overline{b_{jk}^2}, \tag{2.3}$$

$$\begin{aligned}\langle\Omega^4\rangle=&\frac{1}{9}(\gamma MV)^4\Big[\Big(4\langle m_z^4\rangle_0+\frac{\langle m_z^2\rangle_0}{\langle m_x^2\rangle_0}\langle m_x^2m_z^2\rangle_0+\langle m_x^2\rangle_0\\&+3\langle m_x^2m_z^2\rangle_0\Big)\sum_k\overline{b_{jk}^4}+(13\langle m_z^2\rangle_0^2+4\langle m_x^2\rangle_0^2+4\langle m_z^2\rangle_0\langle m_x^2\rangle_0)\sum_{k,p\neq}\overline{b_{jk}^2b_{jp}^2}\\&+(9\langle m_z^2\rangle_0^2-3\langle m_x^2\rangle_0^2+4L\langle m_z^3\rangle_0)\sum_{k,p\neq}\overline{b_{jk}^2b_{jp}b_{kp}}.\end{aligned}$$

Here

$$\begin{aligned}\sum_k\overline{b_{jk}^2}&=\frac{1}{(V_c-V)^{N-1}}\int_{V_c-V}\sum_k b_{jk}^2dx_{j1}dx_{j2}\ldots dx_{jN}\\&=\frac{N-1}{V-V}\int_{V_c-V}b_{j1}^2dx_{j1}\approx\frac{16}{5}\pi^2\frac{c}{V^2},\\ \sum_k\overline{b_{jk}^4}&\approx\frac{256}{35}\pi^4\frac{c}{V^4},\end{aligned} \tag{2.4}$$

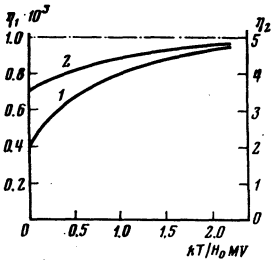

FIG. 2. Temperature dependence of the resonance-line form coefficient η for two volume concentrations c of the ferromagnetic particles. Curve 1—$c=10^{-3}$, left-hand scale; curve 2—$c=0.2$, right-hand scale.

$$\sum_{k,p\neq s}\overline{b_{ik}^2 b_{ip}^2}=\frac{1}{(V_c-V)^{N-2}}\int_{V_c-V}\sum_{k,p\neq s} b_{ik}^2 b_{ip}^2 dx_{i1}dx_{i2}\ldots dx_{iN}$$
$$-\left(\frac{N-2}{V_c-V}\right)^2\int_{V_c-V} b_{i1}^2 b_{i2}^2 dx_{i1}dx_{i2}\approx\frac{256}{25}\pi^4\frac{c^2}{V^4},$$
$$\sum_{k,p\neq s}\overline{b_{ik}^2 b_{ip}b_{kp}}\approx 5\pi^4\frac{c^2}{V^4}.$$

We consider now the temperature dependence of the ratio $\eta=\langle\Omega^4\rangle/\langle\Omega^2\rangle^2$, which characterizes the shape of the resonance line. At volume concentrations $c=NV/V_c\ll 1$ we have approximately

$$\eta\approx\frac{5}{63}\frac{1}{c\langle m_z^2\rangle_0^2}\Big(4\langle m_z^4\rangle_0+\frac{\langle m_z^2\rangle_0}{\langle m_z^2\rangle_0}\langle m_x^2 m_z^2\rangle_0$$
$$+\langle m_z^2\rangle_0+3\langle m_x^2 m_z^2\rangle_0\Big). \qquad (2.5)$$

A plot of this expression for two values of c is shown in Fig. 2. It is seen that at very small volume concentrations of the ferromagnetic particles ($c\sim 10^{-3}$) the line is a quasi-Lorentz curve with $\eta\gg 3$ in the entire temperature interval.

With increasing volume concentration, the line shape changes from quasi-Lorentzian at high temperatures and small particle dimensions to Gaussian at low temperatures and large particle dimensions. At $c\sim 0.1$, the line shape is close to Gaussian in the entire temperature interval, since η is close to 3 ($\eta=3$ for a Gaussian).

The resonance line width $\Delta\Omega$ normalized at $T=0$ is given for a quasi-Lorentzian curve by[8]

$$\Delta\Omega=\frac{\pi}{\sqrt{3}}\langle\Omega^2\rangle^{1/2}\left(\frac{\langle\Omega^2\rangle^2}{\langle\Omega^4\rangle}\right)^{1/2}\langle m_z\rangle_0, \qquad (2.6)$$

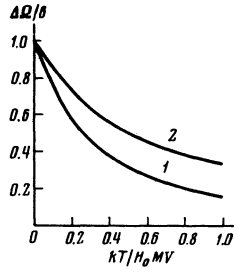

FIG. 3. Temperature dependence of the normalized width of the resonance line for a quasi-Lorentzian line (curve 1) and a Gaussian (curve 2) line.

and for a Gaussian curve by

$$\Delta\Omega=2(2\ln 2)^{1/2}\langle\Omega^2\rangle^{1/2}\langle m_z\rangle_0^{1/2}. \qquad (2.7)$$

Substituting in these formulas the expressions (2.3) for the moments, we obtain for the quasi-Lorentzian curve

$$\Delta\Omega=b\langle m_z\rangle_0\langle m_z^2\rangle_0(5\langle m_z^2\rangle_0)^{1/2}$$
$$\times\left[0.5+\langle m_z^2\rangle_0+2.5\langle m_z^4\rangle_0+\frac{\langle m_z^2\rangle_0(\langle m_z^2\rangle_0-\langle m_z^4\rangle_0)}{1-\langle m_z^2\rangle_0}\right]^{-1/2}, \qquad (2.8)$$

where

$$b=\Delta\Omega|_{T=0}=\sqrt{21/20}\pi^2 c\gamma M,$$

and for the Gaussian curve

$$\Delta\Omega=(0.4\ln 2)^{1/2}\pi\gamma M\sqrt{c}(\langle m_z\rangle_0\langle m_z^2\rangle_0)^{1/2}. \qquad (2.9)$$

The temperature dependences of curves (2.8) and (2.9) are shown in Fig. 3. It is seen that with increasing temperature the line width due to the magneto-dipole interaction decreases. The decrease of the influence of the magneto-dipole interaction is due to averaging of the latter when the magnetization vector executes random temperature oscillations; this decrease is faster the smaller the concentration of the ferromagnetic particles.

For the line width due to the internal relaxation, the opposite relation was obtained,[2,3] namely, an increase of the line width with increasing temperature like

$$\Delta\omega=a(L^{-1}-\sigma^{-1}), \qquad (2.10)$$

where $a=2\xi\omega_0$ is the line width at $T=0$ and ξ is the relaxation parameter in the Landau–Lifshitz equation.

It is difficult to solve the problem with allowance for both the internal relaxation and the magnetodipole interaction, since the method of moments is not applicable to a pure Lorentzian, to which the internal relaxation leads. We confine ourselves therefore to a comparison of the results (2.8) and (2.10), which were independently obtained.

The easiest to investigate is the case of low temperatures, when approximate expressions can be written for (2.8) and (2.10)

$$\Delta\Omega\approx b(1-{}^{11}/_{5}\sigma^{-1}),\quad \Delta\omega\approx a(1+\sigma^{-2}). \qquad (2.11)$$

Assuming that the total line width is approximately described by the expression $\Delta\Omega+\Delta\omega$, we see that it has a minimum at a temperature T_0 given by the rela-

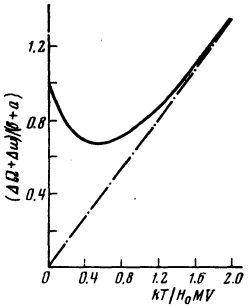

FIG. 4. Temperature dependence of the normalized summary width of the resonance line (solid curve) for $c=10^{-3}$; dash—dot—asymptote of solid curve.

tion

$$a/b \approx \sigma_0 = H_0 MV/kT_0. \quad (2.12)$$

In order for expressions (2.11), from which this equation was obtained, to be valid it is necessary to satisfy the condition $\sigma_0 \gg 1$. The minimum lies therefore in the region of low temperatures (or large values of V) only if $b \ll a$. At other values of the parameters, a minimum also exists and can be obtained graphically. Thus, Fig. 4 shows a plot of the normalized line width $(\Delta\Omega + \Delta\omega)/(b+a)$ at $b = 2a$. It is seen that the minimum is located in this case at $\sigma_0 = 2$. The temperature T_0 corresponding to the minimum line width separates the region where the magnetodipole relaxation mechanism predominates ($T < T_0$) from the region of predominance of the internal relaxation mechanisms ($T > T_0$).

[1]Yu. L. Raĭkher and M. I. Shliomis, Zh. Eksp. Teor. Fiz. **67**, 1060 (1974) [Sov. Phys. JETP **40**, 526 (1975)].
[2]R. S. Gekht, V. A. Ignatchenko, Yu. L. Raĭkher, and M. I. Shliomis, Zh. Eksp. Teor. Fiz. **70**, 1300 (1976) [Sov. Phys. JETP **43**, 677 (1976)].
[3]V. A. Ignatchenko and R. S. Gekht, Zh. Eksp. Teor. Fiz. **73**, 632 (1977) [Sov. Phys. JETP **46**, 331 (1977)].
[4]L. Landau and E. Lifshitz, Phys. Z. Sowjetunion 8, 153 (1935) [included in the Collected Works of L. D. Landau].
[5]J. H. Van Vleck, Phys. Rev. **74**, 1168 (1948).
[6]H. Suhl, Phys. Rev. **97**, 555 (1955).
[7]R. Kubo, J. Phys. Soc. Jpn. **12**, 570 (1957).
[8]A. Abragam, Principles of Nuclear Magnetism, Oxford, 1961.
[9]M. McMillan and W. Opechowski, Can. J. Phys. **38**, 1168 (1960).

Translated by J. G. Adashko

Electroconvective flow in nematics. Velocities and deformation

A. A. Razumov and E. A. Kirsanov

State University, Ivanovo

(Submitted 7 June 1978)
Zh. Eksp. Teor. Fiz. **76**, 175–180 (January 1979)

The optical parameters, orientational deformation angles, and velocities of electroconvective flow in nematic liquid crystals were determined. Finite values of the velocities, independent of the electric field frequency, were obtained at the instability threshold.

PACS numbers: 61.30.Gd, 66.10.Ed

INTRODUCTION

Electroconvective flow in nematic liquid crystals is due to the motion of an electric space charge under the action of an external static or alternating field. This flow is associated with an electrohydrodynamic instability described in several papers.[1,2] The experimental situation is usually as follows.

A nematic crystal is in a flat cell between glass plates coated with transparent and electrically conducting films. The long axes of the nematic molecules are oriented parallel to the glass plates along the rubbing axis x. When a threshold voltage V_{th} is reached in the conduction regime,[3] a system of parallel vortex tubes, oriented along the y axis, i.e., at right-angles to the rubbing direction, appears in the nematic. The existence of a velocity gradient results in a hydrodynamic displacement and corresponding deformation of the orientations of the long axes of the molecules. The maximum angle of inclination H_0 of the molecules is observed at the center of a vortex tube; the molecules remain parallel to the cell walls in the regions of ascending and descending fluxes (along the line of sight). Reorientation of the long axes of the molecules results in a periodic variation of the refractive index along the x axis. This produces a refractive-index gradient which can focus light whose electric vector is parallel to the x axis.[4] The focal lines observed in a polarizing microscope are known as the Williams domains.[5]

EXPERIMENTAL RESULTS

We determined experimentally the parameters of vortex tubes (domains). We observed experimentally[6] a difference between the focal lengths above the regions of ascending and descending fluxes (Fig. 1a). The asymmetry of the optical pattern was due to inequivalence of the convex and concave parts of the pattern of orientation lines, as viewed relative to the light beam vector. A beam passing at an angle to the local optic axis (coinciding, in our case, with the orientation line) became bent, which was not allowed for in the purely gradient "quasiisotropic" model.[4] Our measurements revealed a considerable difference between the upper, average, and lower focal lengths f_u, f_{av}, and f_l, as well as alternation of the lengths between the lower focal lines l and $\lambda - l$ for samples of n-azoxyanisole (PAA) and n-methoxybenzylidene-n'-butylaniline (MBBA) in a wide range of voltages and frequencies. Thus, the optical pattern made it possible to determine unambiguously the ascending and descending (relative to the vertical light beam) parts of the flow of the liquid, as well as

0038-5646/79/010089-04$02.40