at least one of the eigenvalues E_{n_0} must be greater than zero, since

$$\phi=\sum_n a_n\psi_n, \quad \sum_n|a_n|^2=1,$$
$$\phi K\phi=\lambda=\sum_n|a_n|^2E_n>1.$$

Since K effectively decreases continuously with increasing temperature, we expect the eigenvalues to decrease continuously as the temperature increases. Thus, if $E_{n_0}(T=0)>1$, there will be a temperature T_0 at which $E_{n_0}(T_0)=1$. At this temperature $\psi_{n_0}(T_0)$ satisfies (B4′).

If we take ϕ as

$$\phi(\mathbf{p})=[\ln(2/\mu)]^{-1/2}\frac{1}{p}\left[\frac{-\mathcal{P}(\mathbf{p},\mathbf{q})}{\Delta E(\mathbf{p},\mathbf{q})}\frac{8\pi^2}{m}\right]^{1/2}, \quad \mu p_F\leq p\leq e\mu p_F$$
$$=0, \quad p\leq\mu p_F, \quad p\geq e\mu p_F, \tag{B5}$$

it is easily seen that $\phi K\phi$ is proportional to $\ln\mu$ for sufficiently small μ. This completes the proof.

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Quantum Theory of Magnetic-Resonance Line Shape in a Rigid Lattice*

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The autocorrelation function of the transversal magnetic moment is investigated in the presence of dipole-dipole interaction. For that purpose an integrodifferential equation is derived which is exact in all orders of the perturbation. The resulting equation is simplified with the aid of a linearity and Gaussian assumption and is solved numerically. Good agreement is thereby found with the experimental situation. In particular, it is shown that the physical nature of the oscillations found in the free-induction-decay experiments of Lowe and Norberg can be understood as a precession around an average internal magnetic field.

I. INTRODUCTION

IN the calculations of the resonance linewidth in paramagnetic systems with dipole-dipole interaction one in general makes use of the Gaussian assumption for the line shape.[1,2] This has clearly the advantage that the method of moments can be applied in a simple way. On the other hand, it is experimentally demonstrated by Lowe and Norberg[3] using the free-induction-decay method, that the decay curves are of an oscillatory nature (in contrast to the monotonic decrease of a Gaussian function).

The autocorrelation function $\Phi(t)$ of the transversal magnetic moment, which essentially determines the decay curves, has also been evaluated by Lowe and Norberg. Their calculation is based on a power-series expansion of the form

$$\Phi(t)=\sum_{n=0}^{\infty}t^nF_n(t), \tag{1}$$

in lowest order of which the Ising and exchange parts of the dipole-dipole interaction are assumed to commute. Because of the mathematical complications only the terms up to $n=4$ have been calculated. Remarkable agreement is thereby found with the experimental situation. It is, however, not clear from the theoretical point of view whether the higher order terms can be neglected.

The present paper is devoted to the study of the autocorrelation function within the framework of a formalism which has been developed previously[4] and which is based on a many-particle treatment of the spin system. From the present treatment the physical mechanism of the oscillatory behavior in time can, at least qualitatively, be understood.

We first shall be concerned with a derivation of an integrodifferential equation for the autocorrelation function with the use of a power-series expansion in terms of the Ising part of the interaction Hamiltonian. Subsequently, the resulting equation is simplified with the aid of a linearity condition. Finally, it is solved numerically and a comparison is made with experiment.

II. FORMULATION OF THE PROBLEM

In this section we briefly recall some details for further reference. Consider for definiteness a crystal of identical paramagnetic ions with spin $\frac{1}{2}$ which is placed in a large external magnetic field **H**. The field **H** is supposed to be along the z axis. Furthermore, let us confine ourselves to the situation that we may neglect

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[1] J. H. Van Vleck, Phys. Rev. **74**, 1168 (1948).
[2] B. Herzog and E. L. Hahn, Phys. Rev. **103**, 148 (1956).
[3] I. J. Lowe and R. E. Norberg, Phys. Rev. **107**, 46 (1957).
[4] J. A. Tjon, Physica **30**, 1341 (1964) (hereafter to be referred to as I).

the relative motions of the ions, that is the spin-lattice relaxation time is taken to be infinite.

In the CW experiments one is interested in the linear response coefficient of the spin system to a small rf magnetic field with frequency ω along the x axis. This coefficient is given by the so-called complex susceptibility $\chi(\omega)$. As is well known, $\chi(\omega)$ is simply related in the high-temperature limit to the autocorrelation function $\Phi(t)$ in the following way (see, for example, Refs. 5 and 6)

$$\chi(\omega)=-\chi_0\int_0^\infty \frac{d\Phi(t)}{dt}e^{-i\omega t}\,dt, \tag{2}$$

where χ_0 denotes the static susceptibility. $\Phi(t)$ is defined according to

$$\Phi(t)=\mathrm{Tr}\{e^{i\mathcal{H}t}S_1e^{-i\mathcal{H}t}S_1\}/\mathrm{Tr}S_1^2. \tag{3}$$

For convenience we have used units in which $\hbar=1$. In (3) S_1 represents the x component of the total spin operator and $\mathcal{H}$ is the total Hamiltonian of the system, i.e.,

$$\mathcal{H}=\mathcal{H}_z+\mathcal{H}'',$$

where $\mathcal{H}_z$ designates the Zeeman energy and $\mathcal{H}''$ describes the interaction between the spins. It can now be shown that in case $\mathbf{H}$ is large the expression (3) in a good approximation can be reduced to

$$\Phi(t)=\cos\omega_L t\,\Phi_0(t), \tag{4}$$

where ω_L is the Larmor frequency and $\Phi_0(t)$ is the reduced autocorrelation function which is defined as

$$\Phi_0(t)=\mathrm{Tr}\{S_1(t)S_1\}/\mathrm{Tr}S_1^2, \tag{5}$$

with

$$S_1(t)=e^{i\mathcal{H}_t t}S_1e^{-i\mathcal{H}_t t}. \tag{6}$$

Here $\mathcal{H}_t$ denotes the truncated Hamiltonian which consists of only the secular part of $\mathcal{H}''$. We may now write the secular part in the following form

$$\mathcal{H}_t=\lambda_1\mathcal{H}_1+\lambda_2\mathcal{H}_2,$$

where $\lambda_1\mathcal{H}_1$ and $\lambda_2\mathcal{H}_2$ are the exchange and Ising parts of the interaction Hamiltonian, respectively:

$$\lambda_1\mathcal{H}_1=\sum_{j<k}a_{jk}\mathbf{S}_j\cdot\mathbf{S}_k,$$

$$\lambda_2\mathcal{H}_2=\sum_{j<k}b_{jk}S_{j3}S_{k3}. \tag{7}$$

In Eq. (7), $\mathbf{S}_j$ means the spin operator of the jth spin.

III. THE INTEGRODIFFERENTIAL EQUATION

According to the previous section the problem of finding the susceptibility is reduced to the determination of the reduced autocorrelation function $\Phi_0(t)$. For that purpose, the method employed in I will be generalized to all orders of the perturbation. The extension is in principle needed because the perturbation, given in this case by $\lambda_2\mathcal{H}_2$, is not small.

Our starting point is the following power-series expansion for Eq. (6), which is obtained on iterating the equations of motion in the interaction representation

$$S_1(t)=\sum_{n=0}^{\infty}(-i\lambda_2)^n\int_0^t dt_n\int_0^{t_n}dt_{n-1}\cdots\int_0^{t_2}dt_1 \times K_n(t-t_1,t-t_2,\cdots,t-t_n), \tag{8}$$

where K_n is given by the nth commutator

$$K_n(\tau_1,\tau_2,\cdots,\tau_n)=[S_1;\mathcal{H}_2'(\tau_1),\mathcal{H}_2'(\tau_2),\cdots,\mathcal{H}_2'(\tau_n)]_n, \tag{9}$$

with

$$\mathcal{H}_2'(\tau_i)=e^{i\lambda_1\mathcal{H}_1\tau_i}\mathcal{H}_2e^{-i\lambda_1\mathcal{H}_1\tau_i}.$$

We next introduce a complete orthonormal set of states $|\alpha\rangle$ which are eigenfunctions simultaneously of $\mathcal{H}_1$ and S_1:

$$\mathcal{H}_1|\alpha\rangle=E(\alpha)|\alpha\rangle,$$
$$S_1|\alpha\rangle=S(\alpha)|\alpha\rangle.$$

An operator A may in general be decomposed into

$$A=A_\mathrm{d}+A_\mathrm{nd}, \tag{10}$$

so that

$$\langle\alpha|A_\mathrm{d}|\alpha'\rangle=\langle\alpha|A|\alpha\rangle\delta_{\alpha\alpha'}.$$

The operators A_d and A_nd are called the diagonal and nondiagonal parts of A, respectively. Furthermore, we shall also need the simple diagonal part of the mth commutator, defined according to

$$[B;\mathcal{H}_2',\mathcal{H}_2',\cdots,\mathcal{H}_2']_{m;\mathrm{sd}} =[[[\cdots[B,\mathcal{H}_2']_\mathrm{nd}\mathcal{H}_2']_\mathrm{nd}\cdots]_\mathrm{nd}\mathcal{H}_2']_\mathrm{d}, \tag{11}$$

where B is an arbitrary operator which is diagonal in the representation $|\alpha\rangle$. Thus the simple diagonal part is called that portion of the mth commutator which is obtained by retaining the nondiagonal part only of each commutator except the last one of which one should keep the diagonal part. In view of the definitions (10) and (11), we may write for the diagonal part of (9)

$$\{K_n(\tau_1,\cdots,\tau_n)\}_\mathrm{d}=[S_1;\mathcal{H}_2'(\tau_1),\cdots,\mathcal{H}_2'(\tau_n)]_{n;\mathrm{sd}} +\sum_{n_1}[[S_1;\cdots\mathcal{H}_2'(\tau_{n_1})]_{n_1;\mathrm{sd}}\cdots\mathcal{H}_2'(\tau_n)]_{n-n_1;\mathrm{sd}} +\sum_{n_1,n_2}[[[S_1;\cdots\mathcal{H}_2'(\tau_{n_1})]_{n_1;\mathrm{sd}}\cdots\mathcal{H}_2'(\tau_{n_1+n_2})]_{n_2;\mathrm{sd}}\cdots \times\mathcal{H}_2'(\tau_n)]_{n-n_1-n_2;\mathrm{sd}}+\cdots. \tag{12}$$

The above separation amounts to decomposing each term of Eq. (8) into parts which asymptotically for large t behave like t^k, where k is the number of simple diagonal parts in the corresponding terms on the right-hand side of Eq. (12).

[5] A. Abragam, *The Principles of Nuclear Magnetism* (Clarendon Press, Oxford, England, 1961).
[6] R. Kubo and K. Tomita, J. Phys. Soc. Japan **9**, 888 (1954).

On substituting (12) into (8) we find for the diagonal part of $S_1(t)$, after an obvious rearrangement of terms,

$$\{S_1(t)\}_{\mathrm{d}}=S_1+\sum_n(-i\lambda_2)^n\int_0^t dt_n\int_0^{t_n}dt_{n-1}\cdots\int_0^{t_2}dt_1 \times[\{S_1(t_1)\}_{\mathrm{d}};\mathcal{H}_2'(t-t_1),\cdots,\mathcal{H}_2'(t-t_n)]_{n;\mathrm{sd}}. \quad (13)$$

Differentiation of Eq. (13) with respect to t and a change of integration variables yield, instead of Eq. (13),

$$\frac{d}{dt}\{S_1(t)\}_{\mathrm{d}}=\sum_n(-i\lambda_2)^n\int_0^t dt_1\int_0^{t_1}dt_2\cdots\int_0^{t_{n-2}}dt_{n-1} \times[\{S_1(t-t_1)\}_{\mathrm{d}};\mathcal{H}_2'(t_1),\cdots,\mathcal{H}_2'(t_{n-1}),\mathcal{H}_2'(0)]_{n;\mathrm{sd}}. \quad (14)$$

At this point it is convenient to introduce the Hilbert space $\mathfrak{L}$ of linear operators which are diagonal in the representation $|\alpha\rangle$. Similarly, as in I, we may define in this new Hilbert space the linear operators f_n in the following way:

$$f_n(t_1,\cdots,t_{n-1})A=[A;\mathcal{H}_2'(t_1),\cdots,\mathcal{H}_2'(t_{n-1}),\mathcal{H}_2'(0)]_{n;\mathrm{sd}}, \quad (15)$$

where A is in $\mathfrak{L}$. With this the result (14) can be written in a very compact form. We have

$$\frac{d}{dt}\{S_1(t)\}_{\mathrm{d}}=\int_0^t f(\tau)\{S_1(t-\tau)\}_{\mathrm{d}}d\tau, \quad (16)$$

with

$$f(\tau)=\sum_n(-i\lambda_2)^n\int_0^\tau d\tau_1\int_0^{\tau_1}d\tau_2\cdots \times\int_0^{\tau_{n-3}}d\tau_{n-2}f_n(\tau,\tau_1,\cdots,\tau_{n-2}). \quad (17)$$

In addition to Eq. (16) we also have the initial condition

$$\{S_1(0)\}_{\mathrm{d}}=S_1.$$

For completeness, although it will not be needed here, we also give the solution to the nondiagonal part of $S_1(t)$. It is expressed in terms of the diagonal part of $S_1(t)$ according to

$$\{S_1(t)\}_{\mathrm{nd}}=\sum_n^\infty(-i\lambda_2)^n\int_0^t dt_1\int_0^{t_1}dt_2\cdots\int_0^{t_{n-1}}dt_n \times[[\cdots[[\{S_1(t-t_1)\}_{\mathrm{d}};\mathcal{H}_2'(t_2)]_{\mathrm{nd}} \times\mathcal{H}_2'(t_2)]_{\mathrm{nd}}\cdots]_{\mathrm{nd}}\mathcal{H}_2'(t_n)]_{\mathrm{nd}}. \quad (18)$$

In order to discuss the solution of Eq. (16) in detail this equation should be simplified considerably. For that purpose we make use of a linearity assumption. It can be formulated as

$$f(\tau)\{S_1(t-\tau)\}_{\mathrm{d}}=\alpha(\tau)\{S_1(t-\tau)\}_{\mathrm{d}}, \quad (19)$$

where $\alpha(\tau)$ is a c number. $\alpha(\tau)$ can easily be found on multiplying Eq. (19) by S_1 and taking the trace at $t=\tau$. So we find

$$\alpha(\tau)=\mathrm{Tr}\{S_1f(\tau)S_1\}/\mathrm{Tr}S_1^2. \quad (20)$$

With the aid of Eq. (16) together with the linearity condition (19) we obtain the following simple equation for $\Phi_0(t)$

$$\frac{d\Phi_0(t)}{dt}=\int_0^t d\tau\,\alpha(\tau)\Phi_0(t-\tau), \quad (21)$$

with the initial condition

$$\Phi_0(0)=1.$$

The nature of the approximation (19) can be seen on examining Eq. (21) in the following two cases. Firstly, we may study the weak-coupling limit, i.e., $\lambda_2\to 0$, $t\to\infty$ so that $\lambda_2^2t\to$ constant. This has been done in detail in I. As a solution to Eq. (21) we find a pure exponential decay,

$$\Phi_0(t)=e^{-\gamma t},$$

with

$$\gamma=\lambda_2^2\int_0^\infty d\tau\mathrm{Tr}\{[S_1;\mathcal{H}_2'(\tau),\mathcal{H}_2]_2S_1\}/\mathrm{Tr}S_1^2.$$

The second case we may consider is the strong-coupling limit, i.e., $\lambda_1\to 0$. The operators (15) are then independent of their arguments. If we furthermore restrict ourselves to sufficiently small t, we find that Eq. (21) reduces to

$$\frac{d\Phi_0(t)}{dt}=-\omega_0^2\int_0^t\Phi_0(\tau)\,d\tau, \quad (22)$$

where ω_0^2 is the second moment of the reduced autocorrelation function $\Phi_0(t)$. It is given by

$$\omega_0^2=-\lambda_2^2\mathrm{Tr}\{[S_1,\mathcal{H}_2]^2\}/\mathrm{Tr}S_1^2. \quad (23)$$

The solution of (22) is simply given by

$$\Phi_0(t)=\cos\omega_0t. \quad (24)$$

On the other hand, $\Phi_0(t)$ can also be calculated exactly in this case. One obtains

$$\Phi_{0,\mathrm{exact}}=\prod_k{}'\cos(b_{jk}t/2),$$

where the prime means that $j\neq k$. Hence, we see that in the strong-coupling limit and for small t the linearity condition (19) simply amounts to replacing the exact solution by one single oscillation. Physically it corresponds to the approximation that the fields the individual spin feels from the neighboring spins are being replaced by one average field around which the individual spin precesses.

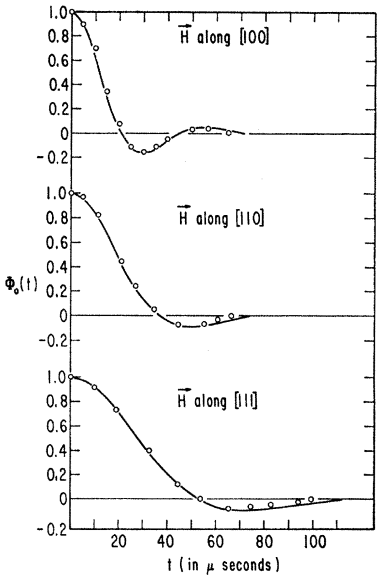

FIG. 1. The free-induction decays as a function of time for several crystal orientations. The circles are some of the experimental points and the full curves represent our results.

In conclusion we should point out that from the above considerations it is obvious that the solution of Eq. (21) in general may exhibit oscillations in the decay. This effect has also been found for continuum systems in the study of the generalized master equation of Van Hove.[7]

IV. COMPARISON WITH THE EXPERIMENTAL SITUATION

Let us now consider the general solution of Eq. (21). It can readily be obtained in a closed form utilizing Laplace transformations. So we find

$$\hat{\Phi}_0(p)=(p-\hat{\alpha}(p))^{-1}, \quad \mathrm{Re}p\geq 0, \tag{25}$$

where

$$\hat{\Phi}_0(p)=\int_0^\infty dt\, e^{-pt}\Phi_0(t),$$

$$\hat{\alpha}(p)=\int_0^\infty dt\, e^{-pt}\alpha(t).$$

As a consequence, we can in view of Eqs. (2) and (4) immediately write down for the susceptibility

$$\chi(\omega)=\chi_0[1-\tfrac{1}{2}i\omega\{\hat{\Phi}_0(i(\omega-\omega_L))+\hat{\Phi}_0(i(\omega+\omega_L))\}]. \tag{26}$$

The measurements of the magnetic-resonance line shapes have been carried out on CaF_2 crystals.[3,8] They have the main advantage that there is only pure dipole-dipole interaction present. Hence, we have

$$-3a_{jk}=b_{jk}=\tfrac{3}{2}\gamma^2(1-3\cos^2\theta_{jk})/r_{jk}^3, \tag{27}$$

where r_{jk} denotes the distance between the jth and kth spin and θ_{jk} the angle between $\mathbf{r}_{jk}$ and the z axis. Furthermore, γ is the gryomagnetic ratio. In order to make a definite comparison with the experiments, we, for simplicity, make the more or less standard type of Gaussian assumption; that is, $\alpha(t)$ is taken to be of the form

$$\alpha(t)=-Ae^{-Bt^2}. \tag{28}$$

The coefficients A and B can then in principle be computed from the first two even moments of $\Phi_0(t)$. The moments M_n of $\Phi_0(t)$, which are defined as (the odd moments vanish)

$$M_{2n}=(-1)^n\left(\frac{d^{2n}}{dt^{2n}}\Phi_0(t)\right)_{t=0},$$

can easily be computed from Eq. (21). On differentiating Eq. (21) repeatedly and taking $t=0$ we find the recurrence relation

$$M_{2n}=\sum_{k=1}^{n}(-1)^k\alpha^{(2(k-1))}(0)M_{2(n-k)}, \tag{29}$$

where

$$\alpha^{(n)}(0)=\left(\frac{d^n}{dt^n}\alpha(t)\right)_{t=0}.$$

In particular, we obtain from Eq. (29) in view of Eq. (28)

$$M_2=A,$$
$$M_4=A^2+2AB. \tag{30}$$

The function $\Phi_0(t)$ may of course be determined by merely taking the inverse Laplace transform of Eq. (25). Instead, however, we have directly solved Eq. (21) numerically. The coefficients A and B can be obtained from Eq. (30), since M_2 and M_4 can be computed explicitly. These moments have in fact been evaluated in the classical paper of Van Vleck.[1] Hence, the solution of Eq. (21) is uniquely determined in terms of known microscopic quantities, and there are therefore no arbitrary parameters involved in Eq. (21). In comparing Eq. (21) with experiment we have adopted the following procedure. $\Phi_0(t)$ was determined from Eqs. (21) and (28), thereby using the theoretical values of A and B which are obtained from Van Vleck's results. Subsequently A and B were varied somewhat around their theoretical values in order to find the best fit to the experiments. The results are shown in Fig. 1 and Table I. The round circles in Fig. 1 denote some of the experi-

TABLE I. The relevant quantities belonging to the several curves of Fig. 1. In the last two columns the results of Van Vleck[a] are given for the two moments.

Direction of H	A_2 (G²)	B_2 (G²)	$M_2^{\frac{1}{2}}$ (G)	$M_4^{\frac{1}{4}}$ (G)	$M_2^{\frac{1}{2}a}$ (G)	$M_4^{\frac{1}{4}a}$ (G)
[100]	11.6	7.6	3.41	4.18	3.6	4.31
[110]	4.5	3.7	2.12	2.68	2.24	2.73
[111]	2.1	1.7	1.45	1.83	1.53	1.88

[a] See Ref. 8.

[7] L. Van Hove and E. Verboven, Physica **27**, 418 (1961).
[8] C. R. Bruce, Phys. Rev. **107**, 43 (1957).

mental points found from the free-induction-decay and CW experiments. The full curves represent our results. The corresponding values for A and B are given in Table I together with the second and fourth moments computed with the aid of Eq. (30). We have also, for convenience, included the results of the explicit calculations of the two moments by Van Vleck. We see that the agreement is gratifying, especially if we also take the experimentally found values for the moments into consideration (see Ref. 8 for these data).

In conclusion we have also examined the influence of the exchange part of the interaction on the first node τ_1 in $\Phi_0(t)$. This is shown in Fig. 2. Qualitatively, it agrees with what one should expect. For $B=0$ the location of the first node is simply given by

$$\tau_1=\pi/2A^{1/2}. \tag{31}$$

When B gets larger, so does τ_1, so that for very strong exchange interaction one will obtain a pure damping without oscillations. Experimentally one finds approximately, for the position of the first zero,

$$\tau_{1,\text{expt}}\approx 2/M_2^{1/2},$$

which is approximately equal to the result calculated above [Eq. (31)] with $B=0$.

V. DISCUSSION

The reduced autocorrelation function has been studied with a technique which essentially takes care of the divergences in time. In doing so we have been able to derive an integrodifferential equation which is valid to all orders of the perturbation.

In order to make a comparison with experiment we had to make a linearity and Gaussian assumption. Needless to say, in general one should be careful in making the Gaussian hypothesis for $\alpha(t)$. The main reason for using this is that the coefficients in the Gaussian function can be simply determined with the aid of the method of moments. A test for the validity of the Gaussian hypothesis is to compute explicitly the fourth moment of the function $\alpha(t)$. Because of the mathematical complexities we have not done this. However, since the exchange term in the interaction is of the same order of magnitude as the Ising term, one should expect that the fourth moment of Eq. (20) will not differ significantly from that of the Gaussian function. From the above considerations it is obvious that one need not expect a perfect agreement with experiment. It is found that the agreement is reasonable in spite of these rather crude approximations.

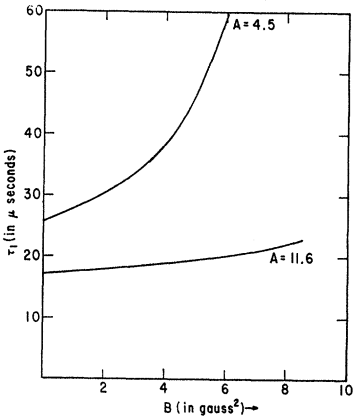

FIG. 2. The dependence on B of the location of the the first zero in the autocorrelation function for a fixed A.

Since the kernel of Eq. (21) reduces in the weak-coupling limit to the lowest order term in λ_2, one may tentatively examine the question in how far the lowest order approximation is also valid in the general case. The kernel in this approximation becomes

$$\alpha(t)=-\lambda_2^2\,\mathrm{Tr}\{[S_1;\mathcal{H}_2'(t),\mathcal{H}_2]S_1\}/\mathrm{Tr}S_1^2. \tag{32}$$

In order to study the above expression we assume it for simplicity to be of a Gaussian form. As a result we obtain

$$\alpha(t)=-A_0e^{-B_0t^2}, \tag{33}$$

with

$$A_0=\langle\nu^2\rangle,$$
$$B_0=\langle\nu^4\rangle/2\langle\nu^2\rangle.$$

Here $\langle\nu^2\rangle$ and $\langle\nu^4\rangle$ are the first two even moments of Eq. (32). They are given by

$$\langle\nu^2\rangle=\lambda_2^2\,\mathrm{Tr}\{[S_1;\mathcal{H}_2,\mathcal{H}_2]S_1\}/\mathrm{Tr}S_1^2=M_2,$$
$$\langle\nu^4\rangle=\lambda_1^2\lambda_2^2\,\mathrm{Tr}\{[S_1;\mathcal{H}_2,\mathcal{H}_1]^2\}/\mathrm{Tr}S_1^2.$$

Hence, we have $A_0=A$. Explicit calculation of $\langle\nu^4\rangle$ for CaF_2, however, shows that the ratio B_0/B is given by about 0.5.