# Behavior of isotropic magnetic materials in two crossed rf magnetic fields

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When two noncollinear magnetic fields with frequencies $\omega_1$ and $\omega_2$ are applied to a magnetic material, a constant moment arises in the direction perpendicular to the plane of these fields, and the magnetization oscillates at the frequencies $\omega_1 \pm \omega_2$ (there is no oscillation at the doubled frequency). These effects are determined in general by ternary dynamic spin correlations. An experimental study of these effects can reveal these correlations, which embody more information about the spin dynamics than does the dynamic susceptibility, which is the property customarily studied. There has been essentially no previous experimental or theoretical study of these correlations. The details of these effects are discussed for two strongly interacting systems: ferromagnets in the critical region above $T_c$ and spin glasses. The type of information which can be extracted from the corresponding experiments is analyzed.

## 1. INTRODUCTION

We use the word "isotropic" here to mean that the magnetic materials under discussion have no preferred direction, e.g., a uniaxial anisotropy or a permanent magnetic moment, either spontaneous or induced by an external field. More specifically, we are interested in cubic ferromagnets in the critical region above the Curie point and in spin glasses. The customary approach in rf experiments on magnetic materials of this type is to measure their dynamic susceptibility $\chi(\omega) = \chi'(\omega) + i\chi''(\omega)$, which describes the linear response to an applied magnetic field (see, for example, Refs. 1–4 and the bibliographies there). There has been considerably less interest in the responses of higher order, which are proportional to odd powers of the applied field (see Refs. 4–6, for example). There has been essentially no discussion of second-order effects which are bilinear in the applied magnetic fields. In general, this is not a surprising situation, since the symmetry of the customary experimental conditions, with a linearly polarized magnetic field applied to the system, usually suppresses such effects (more on this below).

It was shown experimentally as early as 1957, however, that a paramagnetic sample in a rotating magnetic field acquires a static magnetization which is directed along the rotation axis and whose magnitude is proportional to the squared amplitude of the rotating field[1] (Ref. 7 and 8).

In general, when two noncollinear fields with frequencies $\omega_1$ and $\omega_2$ are applied to a sample, oscillations of the magnetization at frequencies $\omega_1 \pm \omega_2$ should arise along the direction of the vector product of these fields. If $\omega_1 = \omega_2$, however, there will be no oscillation at the doubled frequency. We will show below that an experimental study of these effects can reveal genuinely novel and extremely detailed information about the spin systems. Essentially all our knowledge is based on experiments on the dynamic susceptibility, which is a binary spin correlation function. In this new approach, in contrast, it is possible to experimentally study ternary dynamic spin correlations, about which nearly nothing is known. An exceptional case is represented by some recent experiments on neutron scattering in iron[9,10] above $T_c$, but those experiments were carried out at a large momentum transfer, i.e., under conditions far from those discussed in the present paper. It should also be noted that ternary correlations depend on two frequencies and thus contain more information than does the ordinary susceptibility. We will frequently run into situations in which the theoretical assertions can be regarded as at best more or less plausible suggestions. For this reason, experiments on these effects seem extremely promising.

We begin with a theoretical description of these effects. If the relaxation time of the magnetization is long in comparison with the characteristic time for the internal spin motions, we can use the Bloch equation or one of its generalizations (more on this below), as in Refs. 7 and 8. This method is clearly incorrect, however, if these times are comparable in magnitude, as they are, for example, in ferromagnets far from the Curie temperature. In this case the magnetization oscillation amplitude must be expressed in terms of the ternary dynamic spin correlation functions which were introduced in Ref. 11 and regarding which we can draw some definite conclusions based on some particular theory of the spin dynamics.

It should be noted that it is also convenient to speak in terms of spin correlations in the case in which the Bloch equation is applicable, since we can thus immediately draw conclusions from the symmetry of the problem. The structure and symmetry properties of the ternary dynamic correlations are analyzed in detail in Ref. 12.

Using the standard methods, we can write the magnetization which arises in a system in second order in the applied external field $\mathbf{H}(t)$ as

$$m_\alpha(t) = \frac{g\mu}{v_0}\frac{1}{(2\pi)^2}\int d\omega_1\, d\omega_2 \exp\{-i(\omega_1+\omega_2)t\}$$
$$\times \mathcal{F}_{\alpha\rho\varphi}(\omega_1+\omega_2, \omega_1, \omega_2)\,[g\mu H_\rho(\omega_1)]\,[g\mu H_\varphi(\omega_2)], \qquad (1)$$
$$H_\rho(\omega) = \int dt e^{i\omega t} H_\rho(t),$$

where $v_0^{-1}$ is the concentration of magnetic atoms, and $\mathcal{F}_{\alpha\rho\varphi}(\omega_1+\omega_2, \omega_1, \omega_2)$ is an analytic continuation with the

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discrete frequencies of a ternary spin Green's function,

$$\tilde{\mathscr{F}}_{\alpha\rho\varphi}(i\omega, i\omega_1, i\omega_2)\frac{1}{T}\delta_{\omega,\omega_1+\omega_2}=\frac{1}{2}\int_0^{1/T} d\tau_1\, d\tau_2\, d\tau$$

$$\times\exp(i\omega\tau-i\omega_1\tau_1-i\omega_2\tau_2)\langle TS_\alpha(\tau)S_\rho(\tau_1)S_\varphi(\tau_2)\rangle. \quad (2)$$

Here $S$ is the total spin of the system, which is related to its magnetic moment $\mathbf{M}$ by $\mathbf{M}= g\mu v_0^{-1}\mathbf{S}$. The function $\mathscr{F}_{\alpha\rho\varphi}$ is discussed in detail in Ref. 12, so we will simply recall its basic properties here.

1) the function $\mathscr{F}_{\alpha\rho\varphi}(-\omega,\omega_1,\omega_2)$ is a symmetric function of the pairs of arguments $(-\omega,\alpha)$, $(\omega_1,\rho)$, and $(\omega_2,\varphi)$.

2) Since the spin operators are $t$-odd, we have

$$\mathscr{F}_{\alpha\rho\varphi}(\omega+i\delta,\ \omega_1+i\delta,\ \omega_2+i\delta)$$

$$=-\mathscr{F}_{\alpha\rho\varphi}(-\omega-i\delta,\ -\omega_1-i\delta,\ -\omega_2-i\delta)$$

and, in particular, $\mathscr{F}_{\alpha\rho\varphi}(0,0,0)=0$. As expected, the effects in which we are interested here thus vanish in the static limit, $\omega_1=\omega_2=0$.

3) Since $S_\alpha$ is Hermitian we have

$$\mathscr{F}^*_{\alpha\rho\varphi}(\omega+i\delta,\ \omega_1+i\delta,\ \omega_2+i\delta)$$

$$=\mathscr{F}_{\alpha\rho\varphi}(-\omega+i\delta,\ -\omega_1+i\delta,\ -\omega_2+i\delta).$$

4) In accordance with the general principles of the spin diagram technique[13] we can write

$$\mathscr{F}_{\alpha\rho\varphi}(\omega,\ \omega_1,\ \omega_2)$$

$$=G_{\alpha\alpha'}(\omega)G_{\rho\rho'}(\omega_1)G_{\varphi\varphi'}(\omega_2)\Gamma_{\alpha'\rho'\varphi'}(\omega,\ \omega_1,\ \omega_2), \quad (3)$$

where the functions $G$ are the spin Green's functions, defined in the standard way,[13] and $\Gamma$ is the three-spin vertex part. The Green's function is related to the magnetic susceptibility $\tilde{\chi}$, which describes the reaction to an external magnetic field, by[11]

$$\tilde{\chi}_{\alpha\beta}(\omega)=(g\mu)^2 v_0^{-1}G_{\alpha\beta}(\omega). \quad (4)$$

Since $\tilde{\chi}$ depends on the shape of the object, $G$ has the same dependence. On the other hand, the perturbation-theory series for $\Gamma$ is constructed in such a way that small distances are important in all the integrals, so that $\Gamma$ may not depend on the shape of the sample (see Ref. 11 for some corresponding arguments). Since the vertex $\Gamma_{\alpha\rho\varphi}$ is a pseudo-scalar, it is proportional to $\varepsilon_{\alpha\rho\varphi}$ and can be written conveniently as

$$\Gamma_{\alpha\rho\varphi}(\omega,\ \omega_1,\ \omega_2)=\left[\frac{(g\mu)^2}{v_0}\right]^2\varepsilon_{\alpha\rho\varphi}\Gamma(\omega,\ \omega_1,\ \omega_2). \quad (5)$$

According to (1) and (5), the vector $\mathbf{m}$ is thus directed along the vector product of the magnetizations induced by the field $\mathbf{H}$, i.e., along

$$[\mathbf{M}(\omega_1)\times\mathbf{M}(\omega_2)],\quad M_\alpha(\omega)=\tilde{\chi}_{\alpha\beta}(\omega)H_\beta(\omega).$$

We also recall that for an ellipsoid, in the coordinates of its principal axes, we have

$$\tilde{\chi}_i(\omega)=\chi(\omega)\,[1+4\pi N_i\chi(\omega)]^{-1},$$

where $N_i$ are the corresponding demagnetization coefficients, and $\chi(\omega)$ is the ordinary susceptibility. Finally, the properties of $\Gamma$ as a function of the frequencies can easily be found from the properties of $\mathscr{F}$ by using $G_{\alpha\beta}(-\omega)=G^*_{\alpha\beta}(\omega)$.

If two sinusoidal external fields are applied to the system,

$$\mathbf{H}_j={}^1/_2(e^{-i\omega_j t}\mathbf{h}_j+e^{i\omega_j t}\mathbf{h}_j^*);\quad j=1,2, \quad (6)$$

then $\mathbf{m}(t)$ goes into oscillation at the two frequencies $\omega_1\pm\omega_2$, according to (1). For these oscillations we easily find the following expression using (3)–(5):

$$m_\alpha^{(\pm)}(t)={}^1/_2 g\mu\,\mathrm{Re}\{e^{-i(\omega_1\pm\omega_2)t}\tilde{\chi}_{\alpha\beta}(\omega_1\pm\omega_2)$$

$$\times[\mathbf{M}_1(\omega_1)\times\mathbf{M}_2(\pm\omega_2)]_\beta\Gamma(\omega_1\pm\omega_2+i\delta,\ \omega_1+i\delta,\ \pm\omega_2+i\delta)\}, \quad (7)$$

where

$$M_{i\alpha}(\omega_i)=\tilde{\chi}_{\alpha\beta}(\omega_i)h_{i\beta}$$

is the magnetization induced in the sample by the field $\mathbf{h}_i$. In deriving these expressions we used the first symmetry property of $\mathscr{F}$, according to which we have

$$\Gamma(\omega_1+\omega_2,\ \omega_1,\ \omega_2)=-\Gamma(\omega_1+\omega_2,\ \omega_2,\ \omega_1).$$

Because of this property we have $\Gamma(2\omega,\omega,\omega)=0$, so that if $\omega_1=\omega_2$ we have ${}^2\mathbf{m}^{(+)}(t)=0$. Furthermore, we have made use of a third property of $\mathscr{F}$. In addition to the oscillations of $\mathbf{m}$ there is generally also a part which does not depend on the time; this part is the sum of the contributions from the fields $\mathbf{H}_1$ and $\mathbf{H}_2$. A sufficient condition for the existence of this part is that there be a single rotating magnetic field $\mathbf{H}$. In this case the expression for the constant magnetization is

$$m_\alpha^{(0)}={}^1/_2 g\mu\tilde{\chi}_{\alpha\beta}(0)\,\mathrm{Re}\{[\mathbf{M}(\omega)\times\mathbf{M}^*(\omega)]_\beta\Gamma(0,\ \omega+i\delta,\ -\omega+i\delta)\}. \quad (8)$$

In the static limit we would have $\Gamma(0,0,0)=0$, so that $m_\alpha^{(0)}$ would vanish in the limit $\omega\to0$, as expected.

If $x$, $y$, and $z$ are the principal axes of the ellipsoid, and if the field is rotating around $z$, so that $\mathbf{h}=\mathbf{h}_x+\mathbf{i}\mathbf{h}_y$, expression (8) simplifies greatly, and we can write

$$m_z^{(0)}=g\mu h_x h_y\tilde{\chi}_z(0)\,\mathrm{Re}[\tilde{\chi}_x(\omega)\tilde{\chi}_y^*(\omega)]\,\mathrm{Im}\,\Gamma(0,\omega+i\delta,-\omega+i\delta). \quad (9)$$

If the degmanetization is insignificant, and we have $\hat{\chi}\approx\chi$, the expression for $m_z^{(0)}$ becomes even simpler:

$$m_z^{(0)}=g\mu h_x h_y\chi(0)\,|\chi(\omega)|^2\,\mathrm{Im}\,\Gamma(0,\omega+i\delta,-\omega+i\delta). \quad (10)$$

In the opposite limit, of a pronounced demagnetization, i.e., $4\pi N_{i\chi}\gg1$ we find from (9)

$$m_z^{(0)}=g\mu\frac{h_x h_y}{(4\pi)^3 N_x N_y N_z}\,\mathrm{Im}\,\Gamma(0,\omega+i\delta,-\omega+i\delta). \quad (11)$$

It is also a simple matter to write expressions analogous to (9)–(11) for the $m_\alpha^{(\pm)}(t)$.

It follows from (7)–(11) that by measuring $\mathbf{m}(t)$ and knowing the complex susceptibility $\chi(\omega)$ we can determine the dynamic three-spin interaction described by the vertex part of $\Gamma$. It is a particularly simple matter to determine $\Gamma(0,\omega+i\delta,\ -\omega+i\delta)$. Making use of the first and third properties of $\mathscr{F}$, we can easily show that

$$\Gamma(0,\ \omega+i\delta,\ -\omega+i\delta)=i\,\mathrm{Im}\,\Gamma(0,\ \omega+i\delta,\ -\omega+i\delta). \quad (12)$$

At this point we leave the general approach and take up some particular cases.

## 2. FERROMAGNETS ABOVE THE CURIE POINT

We have discussed only the case of cubic ferromagnets. Their state susceptibility, as we known, can be described near the Curie temperature $T_c$ by

$$\chi_0=\frac{Z\omega_0}{4\pi T_c(\varkappa a)^{2-\eta}},\qquad \omega_0=4\pi(g\mu)^2 v_0^{-1},\tag{13}$$

where $\varkappa = a^{-1}\tau^\nu$ is the reciprocal radius of the critical fluctuations, $\tau=(T-T_c)/T_c$, $\nu\approx 2/3$ is the critical exponent of the correlation length, $\eta$ is the Fisher exponent [which is small ($\eta<0.1$) and which will be ignored below], $a$ is a distance slightly shorter than the interatomic distance, and $Z\sim 1$. According to Ref. 14, in the critical region above $T_c$ there are two temperature regions with different types of critical dynamics: an exchange region with $4\pi\chi_0\ll 1$ and a dipole region with $4\pi\chi_0\gg 1$.

We begin with the exchange region. In this region, the demagnetization effects associated with dipole forces are inconsequential, and we have $\tilde{\chi}\approx\chi$. The characteristic energy of the critical fluctuations can be written[15]

$$\Omega_e(\varkappa)=T_c(\varkappa a)^{(5-\eta)/2}\approx T_c\tau^{5/3}.$$

Since the exchange forces conserve the total spin, the relaxation of the homogeneous magnetization is caused primarily by the dipole forces, which can be treated by perturbation theory. As a result, the following expression has been derived[16] for the reciprocal homogeneous-relaxation time:

$$\Gamma_0=1/T_0=\gamma\Omega_e(q_0)\left(\frac{q_0}{\varkappa}\right)^{3/2}\propto\tau^{-1},\tag{14}$$

where $\gamma\sim 1$, and $q_0$ is the dipole momentum which was introduced in Ref. 14 and defined by the condition that if $4\pi\chi_0=1$ then $\varkappa=q_0$. For the dynamic susceptibility we have the Lorentz formula

$$\chi(\omega)=\chi_0 i\Gamma_0(\omega+i\Gamma_0)^{-1}.\tag{15}$$

As was shown in Ref. 17, this expression is valid if $\omega\ll\Omega_e(\varkappa)$; the asymptotic behavior of $\chi(\omega)$ at $\omega\gg\Omega_e(\varkappa)$ was derived in the same paper. The corrections to $\chi(\omega)$ at frequencies $\omega\ll\Omega_e(\varkappa)$ were studied in Ref. 18.

It follows from the condition $\Gamma_0\ll\Omega_e(\varkappa)$ that the magnetization motion in a magnetic field at frequencies small in comparison with $\Omega_e(\varkappa)$ can be described by the Boch equation

$$\dot{\mathbf{M}}=g\mu[\mathbf{MH}]-\Gamma_0(\mathbf{M}-\chi_0\mathbf{H}).\tag{16}$$

This equation was used in Ref. 7 to calculate $m_z^{(0)}$. In our case it is a simple matter to use this equation to derive

$$\Gamma(\omega+i\delta,\ \omega_1+i\delta,\ \omega_2+i\delta)={}^1\!/_2 i(\omega_1-\omega_2)(\Gamma_0\chi_0)^{-2}.\tag{17}$$

As a result, for the amplitudes in (7)-(10) we have

$$m=g\mu H^2\frac{\omega_1-\omega_2}{2(\Gamma_0\chi_0)^2}i\chi(\omega_1+\omega_2)\chi(\omega_1)\chi(\omega_2)\sim\chi_0 H\frac{g\mu H(\omega_1-\omega_2)}{\Gamma_0^2},\tag{18}$$

where $H^2=|[\mathbf{h}_1\times\mathbf{h}_2]|$ for $|[\mathbf{h}_1\times\mathbf{h}_2^*]|$. This expression is conveniently rewritten as

$$m=\frac{g\mu}{v_0}(\varkappa a)^{1/2}i\left[\frac{Z(\omega_1-\omega_2)}{2\Omega_e(\varkappa)}\left(\frac{g\mu H}{\Omega_e(\varkappa)}\right)^2\right]\times\left(\frac{\Omega_e}{\Gamma_0}\right)^2 F(\omega_1+\omega_2)F(\omega_1)F(\omega_2),\tag{19}$$

where $F(\omega)=\Gamma_0(-i\omega+\Gamma_0)^{-1}$. The first factor here, $g\mu v_0^{-1}(\varkappa a)^{1/2}$, is a quantity on the order of the magnetic moment in the similarity theory; the expression in square brackets has a zero scaling dimensionality, since the dimensionality of the field in the similarity theory is the same as $\Omega_e(\varkappa)$. The factor $(\Omega_e/\Gamma_0)^2$ is an enhancement factor which arises because the lifetime of the fluctuations of the homogeneous magnetization is much longer than the lifetime of the critical fluctuations of size $\varkappa^{-1}$. Finally, the product of the functions $F$ is a dynamic form factor, equal to unity in the limit of zero frequency and falling off at frequencies above $\Gamma_0$. It follows from (16) that expression (19) holds if $g\mu H\ll\Gamma_0$, i.e., under conditions more stringent than the ordinary condition of a weak field, $g\mu H\ll\Omega_e(\mathbf{k})$. In general, the effects of interest here reach their maximum at $\omega_i\sim\Gamma_0$. Interestingly, in the case of a constant magnetization, with $\omega_2=-\omega_1$, the quantity $m$ has the ordinary scaling dimensionality at $\omega_1\gg\Gamma_0$, and it falls off as $\omega_1^{-1}$. We also note that, if we ignore the dipole forces but assume the field momentum $\mathbf{q}$ to be nonzero, then by using the expression of Ref. 18 for the Green's function in a field we can determine the vertex part of $\Gamma$. The expression for $m$ differs from (19) only in that $\Gamma_0$ is replaced by $Dq^2$, where $D$ is the spin diffusion coefficient and has the correct scaling dimensionality. We thus see again that the nonscaling dimensionality of (19) arises because we are dealing with relaxation processes which are related to a nonconservation of the total spin and which have characteristic times in the exchange region which are long in comparison with those of the dynamic similarity theory. These results are valid at frequencies low in comparison with $\Omega_e(\varkappa)$. At high frequencies, the decrease in $m$ with increasing frequency is much more pronounced. Since the effects of interest here are small in this case, however, we will not pursue this question further.

We turn now to the dipole region, $4\pi\chi_0\gg 1$. In this region, demagnetization effects are important, and we have $\tilde{\chi}\neq\chi$. Furthermore, the reciprocal relaxation time of the homogeneous magnetization is the same as the characteristic energy $\Omega_d(\varkappa)$ of the critical fluctuations,[14] so that the Bloch equation, (16) can no longer be used. The following expression has been derived for the energy $\Omega_d$ in the simplest case (Ref. 14; see also Ref. 19):

$$\Omega_d(\varkappa)=\Omega_e(q_0)(\varkappa/q_0)^2$$

(the "soft" version of the dynamics). The more detailed analysis of Ref. 14, however, leads to the conclusion that the following equation holds at sufficiently small values of $\varkappa$:

$$\Omega_d(\varkappa)=\Omega_e(q_0)(\varkappa/q_0)$$

(the "hard" version). Since we have $\varkappa\ll q_0$ in the dipole region, we have $\Omega_d(\varkappa)\gg\Omega_e(\varkappa)$ in both cases.

Now all the characteristic times are of the same order of magnitude, and the vertex part must be determined from the general properties of vertices in the theory of second-order phase transitions. The dimensionality of the vertex part of $m$th order is determined by (see Ref. 20, for example)

$$\Gamma_n=(\varkappa a)^{2-\eta}(\varkappa a)^{1/2(1+\eta)(2-n)}\varphi_n, \tag{20}$$

where $\varphi_n$ is a function of the momenta and frequencies in $\Gamma_n$, reduced to a zero dimensionality by means of $\varkappa$ and $\Omega(\varkappa)$, respectively, where $\Omega(\varkappa)$ is the characteristic dynamic-similarity energy. In our case we have $n = 3$; using definition (5), we have[11]

$$\Gamma(\omega,\omega_1+i\delta,\omega_2+i\delta)=T_c\left(\frac{4\pi}{\omega_0}\right)^2(\varkappa a)^{3/2}$$
$$\times i\frac{\omega_1-\omega_2}{2\Omega_d(\varkappa)}\varphi\left(\frac{\omega_1+i\delta}{\Omega_d(\varkappa)},\frac{\omega_2+i\delta}{\Omega_a(\varkappa)}\right), \tag{21}$$

where $\varphi$ is a symmetric function of its arguments. It is natural to assume $\varphi(0,0) = \varphi_0 \sim 1$, although, strictly speaking, this result does not follow from our phenomenological theory. Interestingly, if we had used (17) in place of (21), replacing $\Gamma_0$ by $\Omega_d$, we would have found a result smaller by a factor of $\Omega_e(\varkappa)/\Omega_d(\varkappa)$. This would mean that the ternary vertices in the dynamic-similarity theory are negligibly small. From (21) we find the following expression for the amplitudes $m$:

$$\dot m=\frac{g\mu}{v_0}(\varkappa a)^{1/2}i\frac{Z^3(\omega_1-\omega_2)}{2\Omega_d(\varkappa)}\left(\frac{g\mu H}{\Omega_e(\varkappa)}\right)^2\varphi(\omega_1,\omega_2)$$
$$\times\tilde F_z(\omega_1+\omega_2)\tilde F_x(\omega_1)\tilde F_y(\omega_2), \tag{22}$$

where the factors $\tilde F_i = \chi_0^{-1}\chi_i$ reflect the demagnetization. If the demagnetization is pronounced, then we would have $\tilde F_i = (4\pi N_i\chi_0)^{-1} \ll 1$. The first two factors are analogous to corresponding factors in (19). At the boundary between the dipole and exchange regions, where $q_0 \sim \varkappa$, expressions (19) and (22) are of the same order of magnitude. Working from dynamic-similarity considerations, we can determine the asymptotic behavior fo $\Gamma$ in (21) at $\omega_{1,2} \gg \Omega_d(\varkappa)$. This is done in the Appendix. Using the results derived there, we can easily determine the behavior of the amplitudes (22) as functions of $\omega_{1,2}$ and $\tau$ at high frequencies. We should emphasize here that none of these results, particularly in the dipole region, can be regarded as really rigorous. This caveat applies to both the asymptotic behavior of the various properties and the more important assertion about the scaling dimensionality of the ternary vertex which follows forom (20). Consequently, it would be extremely desirable to see an experimental study of these effects, particularly at low frequencies, $\omega \ll \Omega_d(\varkappa)$, since there are experimental indications (see Ref. 4 and the citations there) that the susceptibility behaves anomalously at low frequencies in the dipole region.

We turn now to some questions associated with the possibility of observing second-order effects. Since these effects vanish in the limit $\omega = 0$, we need to work at frequencies at which the ratio $\omega/\Omega(\varkappa)$ is not to small. According to experimental data (see Refs. 1 and 4, for example), the characteristic frequencies of the critical fluctuations in ferromagnets lie in the range $10^7$–$10^9$ Hz. We can expect the effects of interest to reach their maximum in the same frequency range. Furthermore, the third-order effects—or, more precisely, the third harmonic of the applied field—are comparatively easy to observe experimentally.[4,6] At frequencies $\omega \lesssim \Omega(\varkappa)$ we easily find the following estimate of the amplitude of this harmonic, working from (20):

$$m_3\sim\frac{g\mu}{v_0}(\varkappa a)^{1/2}\left(\frac{g\mu H}{\Omega_e(\varkappa)}\right)^3\tilde F^4(0),$$
$$\tilde F(0)=(1+4\pi N\chi_0)^{-1}, \tag{23}$$

where $H$ is the amplitude of the applied external field, and $N$ is the demagnetizing factor along the field direction. This expression is obviously correct in the dipole region, but it also holds in the exchange region, since in this particular case there is no rotation of the induced moment around the field which would intensify the second-order effects in comparison with the normal scaling estimate.[3] It follows from (22) and (23) that at $\omega \sim \Omega_d(\varkappa)$ the third-order effects would seem at first glance to be small in comparison with the second-order effects because of the factor $g\mu H/\Omega_e(\varkappa)$. However, this is not quite the case, because of the demagnetization: In the case of the third harmonic we can use an experimental geometry such that $\tilde F(0) \approx 1$, while the second-order effects two of the three factors $\tilde F$ in (22) are unavoidably small. In the exchange region we obviously do not have to deal with these complexities, and the second-order effects are larger than the third-order effects except at very low frequencies.

### 3. SPIN GLASSES

At present we have nothing approaching satisfactory theory for the dynamic phenomena in spin glasses. Extensive experiments on both electron spin resonances and magnetization at low frequencies have drawn a crude picture of the events. We will describe this picture and then make use of it. An experimental study of these effects would make it possible to test the validity of this picture for describing more-complicated phenomena and (also to determine the parameters of the theory). The primary interaction in spin glasses is usually a sign-changing exchange. It creates correlated motions of the spins in spatial regions small in comparison with the dimensions of the sample. The effects in which we are interested, which are linked with the dynamics of the homogeneous magnetization, occur because of interactions which are weak in comparison with the exchange interactions but which violate total-spin conservation: Dipole forces and the local Dzyaloshinskiĭ-Moriya interaction, for example, are usually discussed (see Ref. 21, for example). When we also note that the susceptibility of spin glasses is usually small ($4\pi\chi_0 \ll 1$), we are led to expect that the dynamics of homogeneous magnetization would be described by phenomenological equations of the Bloch type. Furthermore, it follows from experimental data that the homogeneous relaxation occurs in different ways in the ESR region (see Ref. 22, for example) and in the low-frequency region.[3,23] These two regions are separated by some frequency $\Omega_0$ which is not yet known but which can be bracketed: $10^3$–$10^4$ s$^{-1} < \Omega^0 < 10^8$ s$^{-1}$. We first consider the low-frequency region, $\omega < \Omega_0$. For this region it has been shown experimentally[23,24] that the magnetization increases in accordance with $M_0 + M_1 \ln t$ after the imposition of a weak static field, where $t$ is the observation time. This behavior prevails for times measured in hours. Furthermore, the experimental data on the real and imaginary parts of the susceptibility satisfy $\partial\chi'/\partial \ln\omega = -(2/\pi)\chi''$ quite accurately.[3,23] In an attempt to describe these properties, Lundgren *et al.*[3] have suggested that a spin glass is characterized by a wide range of relaxation times $\tau$ and that the low-frequency susceptibility is an average over these times:

$$\chi(\omega)=\chi_1+\int_{\tau_{min}}^{\tau_{max}}\frac{d\tau\chi_0(\tau)g_\tau}{\tau(1-i\omega_0\tau)},\qquad \int_{\tau_{min}}^{\tau_{max}}\frac{d\tau}{\tau}g_\tau=1, \tag{24}$$

where $\tau_{\min}^{-1}\sim\Omega_0$, $\tau_{\max}$ is the maximum possible relaxation time (on the order of hours or longer), $\chi_0(\tau)$ and $g_\tau$ are slowly varying functions of $\ln\tau$ and may be assumed essentially constant, and, finally, $\chi_1$ is the part fo the susceptibility which is constant at $\omega<\Omega_0$. These properties follow immediately from (24) in the time (or frequency) interval $\tau_{\min} < t = \omega^{-1} < \tau_{\max}$, and

$$\chi'=\chi_1+\frac{2}{\pi}\chi''\ln\frac{1}{\omega\tau_{min}},\qquad \chi''=\chi_0 g_\tau. \tag{25}$$

Adopting this picture, we would naturally suggest that for each relaxation time $\tau$ there is a corresponding Bloch equation in which $\mathbf{M}$ is replaced by $\mathbf{m}=\mathbf{M}-\chi_1\mathbf{H}$:

$$\dot{\mathbf{m}}=g\mu[\mathbf{mH}]-(\mathbf{m}-\chi_0\mathbf{H})/\tau. \tag{26}$$

The final result for an observable quantity is found by taking an average of the solutions of this equation over all the $\tau$ values with weight factors $g_\tau\tau^{-1}$. Since the $\tau$ spectrum is broad, we can always find values of $\tau^{-1}$ which are on the order of $g\mu H$. We thus cannot seek a solution of Eq. (26) in the form of a series in $g\mu H$. An analysis of Eq. (26) in the general case in which two oscillating fields are imposed, and we have $g\mu H \gtrsim \tau^{-1}$, is difficult and goes beyond the scope of the present paper. We will restrict the discussion here to some simple cases.

We assume a single rotating field with a circular polarization. Equation (26) can then be solved exactly, and the magnetization is found to be[7]

$$m_z=\frac{(\chi_0 h)g\mu h\omega}{\omega^2+\tau^{-2}+(g\mu h)^2}, \tag{27}$$

where $h$ is the amplitude of the rotating field.[4] Taking an average of this expression over $\tau$ under the condition $\tau_{\max}^{-1}\ll\omega$, $g\mu h\ll\tau_{\min}^{-1}$, we find

$$m_z=\chi''h\frac{g\mu h\omega}{\omega^2+(g\mu h)^2}\ln\{\tau_{max}[\omega^2+(g\mu h)^2]^{1/2}\}, \tag{28}$$

where, according to (25), $\chi''=\chi_0 g_\tau$. At the maximum, which occurs at $\omega = g\mu h$ we have

$$(m_z)_{max}={}^1/_2\chi''h\ln(\tau_{max}g\mu h). \tag{29}$$

Because of the large logarithm, the value of the magnetization at the maximum is considerably higher than the contribution to the oscillation of the magnetization in the approximation linear in $h$, which stem from the imaginary part of the susceptibility. It is difficult, however, to measure the constant magnetization. Accordingly, in the Appendix we analyze the question of oscillations of the magnetization in the case in which two rotating fields, $\mathbf{H}_1$ and $\mathbf{H}_2$, with frequencies $\omega_1$ and $\omega_2$ which are not greatly different are imposed $\omega_1\approx\omega_2\approx\omega$ and $\Delta\omega=|\omega_1-\omega_2|\ll\omega$. It turns out that the effect is maximized when $h_1=h_2=h$ and $g\mu h=\omega$. In this case we find the following expressions for the constant component of the magnetization and for the first two harmonics, with logarithmic accuracy:

$$\begin{aligned}
m_0&=\chi''h\left\{2\ln(\tau_{max}\Delta\omega)+(1-3^{-1/2})\ln\frac{g\mu h}{\Delta\omega}\right\},\\
m_1&=\chi''h\left\{2\ln(\tau_{max}\Delta\omega)+(3\cdot 2^{1/2}-2)\ln\frac{g\mu h}{\Delta\omega}\right\}\cos(\Delta\omega t),\\
m_2&=-\chi''h\frac{(3^{1/2}-1)^2}{2\cdot 3^{1/2}}\ln\left(\frac{g\mu h}{\Delta\omega}\right)\cos(2\Delta\omega t).
\end{aligned} \tag{30}$$

In contrast with the real part of the susceptibility, (25), these expressions depend on the maximum relaxation time $\tau_{\max}$, as do (28) and (29). An experimental test of these results would reveal whether the concept of a broad spectrum of relaxation times has a profound physical meaning or is useful only for a phenomenological description of the relationship between $\chi'$ and $\chi''$, as in Refs. 3 and 23. Furthermore, it becomes possible to directly determine $\tau_{\max}$ and to study its behavior as a function of the external parameters, in particular, a static external field. Indications that there is such a dependence were found in Ref. 23. A weak static field would evidently not interfere with the existence of the magnetization oscillations in question.

At high frequencies, $\omega\gg\tau_{\min}^{-1}\sim\Omega_0$, the slowly relaxing contribution to the susceptibility [the second term in (24)] is negligibly small, but on the other hand we need to consider the dispersion of the first term. The simplest assumption is that in this region we have a Bloch equation with a single relaxation time $\tau_0<\tau_{\min}$. The expressions for the effects of interest then turn out to be the same as those derived above for the exchange critical region in ferromagnets—(17) and (18) with $\Gamma_0$ replaced by $\tau_0^{-1}$ and $\chi_0$ replaced by $\chi_1$. Experiments on ESR in metals,[22] however, have shown that the spins "sense" the local directions of the anisotropy axes, so that in describing the dynamics of the homogeneous magnetization we must take into account not only its rotation around the magnetic field but also the rotation of the spins from their original position. We do not rule out the possibility that a similar situation prevails in insulating spin glasses. Correspoonding phenomenological equations have been formulated.[21,22,25] For the case of interest here, in which the angle ($\psi$) through which the magnetization rotates is small, these equations can be written

$$\begin{aligned}
\dot{\mathbf{M}}&=g\mu[\mathbf{M}\times\mathbf{H}]-\tau_0^{-1}(\mathbf{M}-\chi_1\mathbf{H})-g\mu K\boldsymbol{\psi},\\
\dot{\boldsymbol{\psi}}&=g\mu(\mathbf{M}\chi_1^{-1}-\mathbf{H})-\tau_1^{-1}\boldsymbol{\psi},
\end{aligned} \tag{31}$$

where the vector $\boldsymbol{\psi}$ is directed perpendicular to the plane of the rotation and has a modulus $\psi$. From these equations we find the following expression for the susceptibility:

$$\chi(\omega)=\frac{\chi_1}{\tau_0}\frac{(-i\omega+\tau_1^{-1}+\tau_0\omega_i^2)}{(-i\omega+\tau_0^{-1})(-i\omega+\tau_1^{-1})+\omega_i^2}, \tag{32}$$

where $\omega_i^2(g\mu)^2K\chi_1^{-1}$. If we ignore the terms $\tau_{0,1}^{-1}$ in (31), we find that there is a resonance at the frequency $\omega_i$. The same resonant frequency can be found from the results of Refs. 21 and 22 if the constant field is zero. According to the data of Ref. 22, for the alloy CuMn the frequency $\omega_i$ corresponds to a field of several kilogauss. The physical reason for the existence of a resonance is that it is not favorable from the energy standpoint for the spins to depart from their local equilibrium positions. The terms containing $\tau_{0,1}^{-1}$ in the denominator in (32) give the resonance a finite width. Working from (32), we can write an expression for the amplitudes (7)–(10) which is analogous to (18):

$$m=g\mu H^2[\chi(\omega_1)-\chi(\omega_2)]$$

$$\times\frac{-i(\omega_1+\omega_2)+\tau_1^{-1}}{[-i(\omega_1+\omega_2)+\tau_0^{-1}][-i(\omega_1+\omega_2)+\tau_1^{-1}]+\omega_i^2}. \tag{33}$$

These effects evidently reach a maximum when one of the frequencies is equal to the resonant frequency. There is no point in discussing this expression in more detail until we have some experimental data to look at.

I wish to thank J. Kötzler for the discussions of questions of the critical dynamics of ferromagnets which inspired this study. I also wish to thank B. P. Toperverg for many discussions and useful comments.

## APPENDIX

### 1. Asymptotic behavior of a ternary vertex

Let us determine the asymptotic properties of a ternary vertex in the dipole region under the condition $\omega_{1,2}\Omega_d(\varkappa)$; for simplicity we restrict the discusssion to frequencies below the limiting dipole frequency

$$\Omega_d(q_0)=\Omega_e(q_0)=T_c(q_0a)^{5/2}.$$

In this frequency range, according to Ref. 17, the susceptibility has the standard Lorentzian form with a width of order $\Omega_d(\varkappa)=\Omega_e(q_0)(\varkappa/q_0)^2$ in the "soft" version of the dynamics. In the "hard" version, the asymptotic behavior of the susceptibility is described by[17]

$$\chi(\omega)=\frac{1}{4\pi}\left(\frac{\omega_0}{T_c}\right)^{1/2}\lambda\left(\frac{i\omega_0}{\omega}\right)^{2-3/2|\alpha|}, \tag{A.1}$$

where $\lambda\sim1$, and $\alpha$ is the heat-capacity exponent. We consider two cases of the asymptotic behavior of the vertex part. In the first case, all three frequencies are large, while in the second we have $\omega_1\approx-\omega_2\approx\omega>\Omega_d(\varkappa)$, but a small frequency difference, $|\omega_1+\omega_2|=\Delta\omega\ll\omega$. We consider the first case first. We find the asymptotic behavior from the standard requirement of similarity theory, according to which the dependence on $\varkappa$ or, in other words, on $\tau$ should disappear in the high-frequency limit. It then follows from (21) that

$$\Gamma(\omega_1+\omega_2,\omega_1,\omega_2)=T_c\left(\frac{4\pi}{\omega_0}\right)^2(q_0a)^{3/2}\left[\frac{\omega_1-\omega_2}{\Omega_e(q_0)}\right]^x f_1\left(\frac{\omega_1}{\omega_2}\right), \tag{A.2}$$

where $x=3/4$ and $3/2$ in the soft and hard versions, respectively, and the function $f_1$ is on the order of unity. This function is constructed in such a manner that it leads to all the symmetry properties of the vertex which were formulated at the beginning of this paper. In the second case, we work from the following arguments (for which we claim nothing approaching a rigorous basis): If one of the momenta $q$ going into an $n$-particle vertex is large in comparison with $\varkappa$ and the other momenta in the static limit, then the dependence on this momentum can be singled out as a factor $q^{1/\nu-1-\eta}\approx q^{1/2}$. This is a consequence of the Polyakov-Kadanoff operator algebra,[20] which has recently found experimental support.[9] If we assume that an analogous factorization, with the same scaling dimensionality, can be carried out when one of the frequencies is high, then we find the following expression by working from (12):

$$\Gamma(\omega_1-\omega_2,\omega_1,-\omega_2)$$

$$=T_c\left(\frac{4\pi}{\omega_0}\right)^2(\varkappa a)\left[\frac{\omega}{\Omega_e(q_0)}\right]^{x/3}f_2\left(\frac{\Delta\omega}{\Omega_d(\varkappa)}\right). \tag{A.3}$$

We would naturally expect to have $f_2(0)=\text{const}$. If this asymptotic behavior does actually occur, then the constant magnetization and the oscillations of the magnetization at the frequency $\Delta_0\ll\Omega_d$ would have a temperature dependence $\varkappa\tilde{\chi}_z=(0)$ by virtue of (22).

### 2. Solution of the Bloch equation

From Eq. (26) we find an integral equation for the magnetization along the $z$ axis for the case of fields $\mathbf{H}_1$ and $\mathbf{H}_2$ with frequencies $\omega_1$ and $\omega_2$, which are circularly polarized in the $(x,y)$ plane:

$$m(t)=m_1(t)-{}^1/_2(g\mu)^2\int_{-\infty}^{\infty}dt_1dt_2G(t-t_1)G(t_1-t_2)$$

$$\times[H_+(t_2)H_-(t_1)+H_-(t_2)H_+(t_1)]m(t_2), \tag{A.4}$$

$$m_1(t)=(i\chi_0g\mu/2\tau)\{(h_1^2[G(-\omega_1)-G(\omega_1)]+h_2^2[G(-\omega_2)-G(\omega_2)])G(0)$$

$$+h_1h_2(e^{i(\omega_1-\omega_2)t}G(\omega_2-\omega_1)[G(-\omega_1)-G(\omega_2)]+\text{c.c})\}, \tag{A.5}$$

where $G(\omega)=(-i\omega+\tau^{-1})^{-1}$. A solution of this equation is

$$m(t)=m_0+\sum_{n\neq0}m_ne^{in(\omega_1-\omega_2)t},\quad m_{-n}=m_n^*. \tag{A.6}$$

The coefficients $m_n$ satisfy the system of equations

$$m_n=m_n^{(1)}+l_{n,n}m_n+l_{n,n-1}m_{n-1}+l_{n,n+1}m_{n+1}, \tag{A.7}$$

where the $m_n^{(1)}$ are given by (A.5). For the coefficients $l_{n,n'}$ we have

$$l_{n,n}=l_{-n,-n}^*=-{}^1/_2(g\mu)^2G(\omega_{21}n)$$

$$\times\{h_1^2[G(-\omega_1+\omega_{21}n)+G(\omega_1+\omega_{21}n)]$$

$$+h_2^2[G(-\omega_2+\omega_{21}n)+G(\omega_2+\omega_{21}n)]\}, \tag{A.8}$$

$$l_{n,n-1}=l_{-n,-n+1}^*=-{}^1/_2(g\mu)^2h_1h_2G(\omega_{21}n)$$

$$\times[G(\omega_2+\omega_{21}n)+G(-\omega_1+\omega_{21}n)].$$

If $\Delta\omega = |\omega_{21}| \ll \tau^{-1}$ the coefficients $l_{n,n'}$ are independent of $n$ up to $n \sim (\Delta\omega\tau)^{-1} = n_0$. At $n \ll n_0$ the amplitudes $m_n$ can evidently be determined from Eqs. (A.7) with coefficients $l_{n,n'}$ in which the quantity $\omega_{21}n$ in (A.8) is replaced by zero. The corresponding expressions are

$$m_n = \frac{\chi_0\omega}{g\mu}\left\{\delta_{n,0} - \frac{(-1-A+[(1+A)^2-4B^2]^{1/2})^n}{(2B)^{n+1}[(1+A)^2-4B^2]^{1/2}}\right\}, \qquad (A.9)$$

where

$$A=(h_1^2+h_2^2)\alpha, \quad B=h_1h_2\alpha; \quad \alpha=(g\mu\tau)^2[1+(\omega\tau)^2]^{-1}.$$

This expression is correct for $0 < n \ll n_0$. If $(\Delta\omega\tau)^{-1} \ll 1$, on the other hand, then Eqs. (A.7) can be solved by perturbation theory, with this quantity used as a small parameter. In the lowest order, $m_0$ and $m_1$ are determined from (A.5). In taking the average over $\tau$ for the first few harmonics we must clearly use expression (A.9) if $\Delta\omega\tau < 1$, and we must ue perturbation theory if $\Delta\omega\tau > 1$; in either limiting case, $m_n$ is independent of $\Delta\omega$. In the region $\Delta\omega\tau \sim 1$ there is a smooth transition from one solution to the other. Since the integrals over $\tau$ ae logarithmic in both of the asympotitic regions, the quantity $\Delta\omega$ effectively enters only the argument of the logarithm. Working from these results, we can easily write an expression for $m_n$ in these regions for abitrary $h_{1,2}$ and for $\omega \sim g\mu h_{1,2}$. We will not reproduce these lengthy expressions here. The values of $m_n$ at the maximum are given in the main text of this paper.

[1)] I wish to thank V. N. Fomichev for bringing this point to my attention.

[2)] Strictly speaking, for $m^{(+)}$ to be equal to zero when the frequencies are equal we do not need this property of $\Gamma$, since even before we use it the difference $\Gamma(\omega_1+\omega_2,\omega_1,\omega_2) - \Gamma(\omega_1+\omega_2,\omega_2,\omega_1)$ enters the expression for $m^{(+)}$.

[3)] If there are three nonparallel fields with different frequencies, there will be oscillations of **m** at combinational frequencies with a gain analogous to that in (19). As the frequencies approach zero, these oscillations disappear.

[4)] Interestingly, in the case of an elliptical polarization oscillations of $m_z$ appear at the doubled frequency with an amplitude proportional to $h_x^4 - h_y^4$ at low fields.


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