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Acoustic susceptibility of some insulating spin-glasses

P. Doussineau (1), A. Levelut (1), M. Matecki (2) and W. Schön (1)

(1) Laboratoire d’Ultrasons (*), Université Pierre et Marie Curie, Tour 13, 4 place Jussieu, 75252 Paris Cedex 05, France
(2) Laboratoire de Chimie Minérale D (*), Université de Rennes-Beaulieu, 35042 Rennes Cedex, France

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Abstract. — High frequency acoustic measurements have been performed in three insulating spin-glasses in the vicinity of the spin freezing temperature $T_c$. The results obtained on a nickel fluorophosphate glass are reported and discussed in detail. They are compared with those obtained on manganese, cobalt and zinc fluorophosphate glasses. A similar behaviour is evidenced in the three magnetic glasses. In particular, for the nickel and the cobalt glasses, the static acoustic susceptibility shows a maximum at $T_c$ while the relaxation time presents no anomaly at this temperature.

1. Introduction.

During a long time the experimental study of spin-glasses (SG) was limited to metallic alloys containing magnetic impurities. CuMn and AuFe are the archetypes of these spin-glasses.

A few years ago, insulating spin-glasses appeared. They are either crystals (such as EuSrS) or magnetic glasses (such as fluorophosphates or aluminosilicates of transition elements) [1]. One of the interesting features of insulating compounds is that they are easily probed by acoustic techniques.

A specificity of acoustic studies is the peculiar form of the coupling between the spins and an elastic strain (quadratic or bilinear in spin operators). Consequently four-spin correlation
functions are measured. In this sense the acoustic techniques are complementary of more classical methods (magnetic susceptibility and magnetization measurements).

Recently we have undertaken a systematic study of fluorophosphate glasses containing magnetic ions of the iron group (manganese, nickel, cobalt). Data concerning \((\text{CoF}_2)_3(\text{BaF}_2)_2(\text{NaPO}_3)_3\) have already been reported [2, 3]. Priority was given to this compound because the effective spin \(S = 1/2\) of \(\text{Co}^{2+}\) ions makes the interpretation easier. These data were examined within the frameworks of the Sherrington-Kirkpatrick model and of a dynamical theory. The effect of the spin-glass transition on the sound attenuation and velocity has been satisfactorily explained. The purpose of the present article is to show that the features observed on the cobalt glass are shared by all the glasses of the series. Indeed the attention is focused on the nickel glass \((\text{NiF}_2)_3(\text{BaF}_2)_2(\text{NaPO}_3)_3\) where the magnetic effects are the most spectacular. In addition, for the sake of comparison some results on the cobalt glass \((\text{CoF}_2)_3(\text{BaF}_2)_2(\text{NaPO}_3)_3\), on the manganese glass \((\text{MnF}_2)_6(\text{BaF}_2)_2(\text{NaPO}_3)_3\), and on the non-magnetic glass \((\text{ZnF}_2)_6(\text{BaF}_2)_2(\text{NaPO}_3)_2\) are recalled [4].

In this manner, nearly all the possible cases are represented. There are glasses with:

i) a non-magnetic ion \((\text{Zn}^{2+})\);

ii) a Kramers ion with an effective spin \(S = 1/2\) \((\text{Co}^{2+})\);

iii) an ion with a spin larger than 1/2, here with \(S = 1\) \((\text{Ni}^{2+})\); and

iv) an S-state ion with a spin \(S = 5/2\) \((\text{Mn}^{2+})\).

These different cases correspond to different spin-phonon couplings. For a Kramers ion with \(S = 1/2\) the only possible mechanism (in the absence of biasing magnetic field) is the Waller coupling (modulation of the spin-spin interaction by the elastic waves). For an ion \(S > 1/2\) such as \(\text{Ni}^{2+}\), the Van Vleck mechanism (modulation of the ligand field which acts on the spin through the spin-orbit coupling) is known to be quite efficient [5, 6]. Finally for an S-state ion (no spin-orbit coupling) the Van Vleck mechanism is only weakly efficient and it is not possible to foresee which mechanism (Waller or Van Vleck) is dominant.

The article is divided into two principal parts. We first report on our experimental data which put in evidence the temperature and frequency variations of the elastic properties of the studied glasses. Then we recall some elements of theory necessary for the interpretation of our data and we deduce the relaxation time and the acoustic susceptibility of our nickel glass. We conclude with a short comparison of the properties of the other glasses of the series.

2. Experiments.

All the materials used in the experiments reported in this article were obtained with the same procedure. Details can be found elsewhere [7].

The samples were prepared for ultrasonic work in form of parallelepipeds, the dimensions of which are typically \(4 \times 4 \times 6 \text{ mm}^3\) with two parallel and flat end faces. Ultrasonic waves were generated by resonant \(\text{LiNbO}_3\) transducers directly bonded on a metallized end face. The frequency range spreads from \(30 \text{ MHz}\) up to \(1500 \text{ MHz}\), and the temperature domain extends from \(0.1 \text{ K}\) up to \(8 \text{ K}\) including the freezing temperature \(T_c\) of the spins near \(2 \text{ K}\) depending on the material.

Standard pulse echo techniques were used to measure the changes of the attenuation and of the velocity of the acoustic waves. The experiments on the Co and Ni compounds were performed with a new experimental set-up using a synthesizer. Consequently the accuracy of the recent measurements was improved by about one order of magnitude as compared to the older measurements on the Mn and Zn compounds [4].

In table I some useful data concerning the four materials are presented: chemical formula, density, sound velocities for longitudinal and transverse polarizations.
Table 1. — Some useful data concerning four glasses of general formula 
$(MF_2)_x(BaF_2)_y(NaPO_3)_z$, where $M$ is Zn, Mn, Ni or Co. The upper part of the table gives the 
composition of the glasses. The intermediate part concerns the elastic properties of the glasses:
 density $\rho$, longitudinal and transverse sound velocities $v_L$ and $v_T$ respectively. The lower part of 
the table gives the TLS parameters used in the calculation of the glassy contribution to the elastic 
constant (see text for more details).

$\text{(MF}_2)_x(BaF_2)_y(NaPO_3)_z$

<table>
<thead>
<tr>
<th>M</th>
<th>Zn</th>
<th>Mn</th>
<th>Ni</th>
<th>Co</th>
</tr>
</thead>
<tbody>
<tr>
<td>$x$</td>
<td>0.60</td>
<td>0.65</td>
<td>0.50</td>
<td>0.50</td>
</tr>
<tr>
<td>$y$</td>
<td>0.20</td>
<td>0.20</td>
<td>0.20</td>
<td>0.20</td>
</tr>
<tr>
<td>$z$</td>
<td>0.20</td>
<td>0.15</td>
<td>0.30</td>
<td>0.30</td>
</tr>
<tr>
<td>$\rho$ (kg m$^{-3}$)</td>
<td>4 240</td>
<td>4 050</td>
<td>4 000</td>
<td>3 880</td>
</tr>
<tr>
<td>$v_L$ (m s$^{-1}$)</td>
<td>4 600</td>
<td>4 700</td>
<td>4 750</td>
<td>4 850</td>
</tr>
<tr>
<td>$v_T$ (m s$^{-1}$)</td>
<td>2 300</td>
<td>2 500</td>
<td>2 550</td>
<td>2 500</td>
</tr>
<tr>
<td>$C_L$ (K$^{-1}$ s$^{-1}$)</td>
<td>$4.5 \times 10^{-4}$</td>
<td>$2.3 \times 10^{-4}$</td>
<td>$2.2 \times 10^{-4}$</td>
<td>$2.6 \times 10^{-4}$</td>
</tr>
<tr>
<td>$C_T$ (K$^{-1}$ s$^{-1}$)</td>
<td>$5.4 \times 10^{-4}$</td>
<td>$4.0 \times 10^{-4}$</td>
<td>$7.5 \times 10^{10}$</td>
<td>$4.4 \times 10^{-4}$</td>
</tr>
<tr>
<td>$K_3$ (K$^{-3}$ s$^{-1}$)</td>
<td>$5.5 \times 10^8$</td>
<td>$2.5 \times 10^{10}$</td>
<td>$7.5 \times 10^8$</td>
<td>$2.2 \times 10^9$</td>
</tr>
<tr>
<td>$\mu$</td>
<td>1/4</td>
<td>3/8</td>
<td>1/4</td>
<td>1/4</td>
</tr>
</tbody>
</table>

The materials are in an amorphous state. Therefore in addition to a possible magnetic 
contribution, the usual acoustic behaviour of glasses is also expected. Figure 1 shows the 
results of the velocity change measurements as functions of the logarithm of the temperature 
for several frequencies in the Ni glass. At the lowest temperatures the well known logarithmic 
increase [8] of the velocity is clearly observed. Around 2 K a negative contribution is 
superposed; at the lowest frequencies a marked minimum is even obtained. We attribute this 
second contribution to magnetic effects near $T_c$. Its magnitude decreases with increasing 
frequency but it is always measurable at 1 556 MHz. The lines in figure 1 are the calculated 
velocity variations only taking the glassy nature of the network into account. Details will be 
given in section 3.2.

Two typical sets of data for the attenuation are reported in figure 2. The magnetic 
contribution is not so obvious as for the velocity data. However it is brought into evidence by 
comparison with the behaviour expected for a non-magnetic glass as given by the lines.

In order to make the comparison between the four materials easier, the velocity data are 
shown at neighbouring frequencies for the Mn, Co, Ni and Zn fluorophosphate glasses in 
figure 3. The data for the Co and Ni glasses are quite similar while, as expected, the Zn glass 
shows a genuine glass velocity variation. The behaviour of the Mn glass appears somewhat 
different because the glassy features are anomalous as discussed in a previous article [4].


3.1 THEORETICAL BACKGROUND. — The purpose of the present section is to find the best 
possible fit between a theory and the magnetic contribution obtained at various frequencies 
on samples containing ions with spins $S \geq 1/2$. This demand is too much constraining. In fact
we have at our disposal either a static theory valid for any spin $S \geq 1/2$ [9] or a dynamical theory only valid for $S = 1/2$ [3].

In this section we begin by a brief summary of the two theories. Then we interpret our experiments on the nickel glass.

3.1.1 Static theory. — Sherrington and Kirkpatrick have proposed for spin-glasses a theory which is simple but however retains many of the observed properties. The model, initially built for Ising spins $S = 1/2$ [10], may be extended to any value $S \geq 1/2$ [11]. The spin-phonon coupling may be incorporated into the model. This leads to a dependence of the mean free energy $F(\varepsilon)$ on the static elastic strain $\varepsilon$. The static elastic constant $c$ is easily deduced:

$$c = c_0 + \frac{1}{\vartheta} \frac{d^2 F}{d\varepsilon^2}$$  \hspace{1cm} (1)

$\vartheta$ is the volume of the sample and $c_0$ is the elastic constant without spins. The form of the elastic constant change $\delta c = c - c_0$ due to the spins depends on the coupling mechanism.
Fig. 2. — Sound attenuation as a function of temperature for longitudinal waves in the magnetic glass \((\text{NiF}_2)_3(\text{BaF}_2)_2(\text{NaPO}_3)_3\). The lines are calculated within the TLS theory. 92 MHz: stars and dashed line; 330 MHz: open circles and full line.

i) For the Van Vleck interaction (which is dominant for \(\text{Ni}^{2+}\) ions) we have found \([9]\):

\[
\frac{\delta c}{c_0} = -\frac{\mathcal{N}G^2 A}{c_0 kT B}
\]

where \(\mathcal{N}\) is the number of spins per unit volume, \(T\) is the temperature, \(k\) is the Boltzmann constant, \(G\) is the spin-phonon coupling defined by \(\mathcal{K}_c = -\sum_i G \varepsilon (S_i^2)\), \(A\) and \(B\) are two combinations of four-spin operator averages which can be written:

\[
A = (r-s) - \beta^2 \mathcal{F}'[(r-s)(3v+s-4u) + 2(w-u)^2]
\]

\[
B = 1 - \frac{1}{2} \beta^2 \mathcal{F}'[(r-s) + 2(3v+s-4u)] + \frac{1}{2} \beta^4 \mathcal{F}'[(r-s)(3v+s-4u) + 2(w-u)^2]
\]
Fig. 3. — Relative sound velocity change as a function of temperature for longitudinal waves in four different glasses whose exact composition is given in table I. The lines are calculated within the TLS theory. Co glass: stars and dashed line. Ni glass: open circles and full line. Mn glass: crosses and dashed-dotted line. Zn glass: plusses and dotted line.

where $\beta = 1/kT$. The averages are (with $S_i = S_i^0$):

\[
\begin{align*}
    r &= \frac{1}{N} \sum_i \langle S_i^4 \rangle, \\
    s &= \frac{1}{N} \sum_i \langle S_i^2 \rangle^2, \\
    u &= \frac{1}{N} \sum_i \langle S_i^2 \rangle \langle S_i \rangle^2, \\
    v &= \frac{1}{N} \sum_i \langle S_i^4 \rangle, \\
    w &= \frac{1}{N} \sum_i \langle S_i^3 \rangle \langle S_i \rangle.
\end{align*}
\]

(4)

They are temperature dependent (except in special cases). The bar indicates an average over the random values of the exchange constants $J_{ij}$ which couple the $N$ spins by pairs; the standard deviation of the $J_{ij}$ distribution is $\bar{J}^2/N$. Evidently one has $N = N^0 \theta$. In equation (2)
the only unknown quantities are the coupling constant $G$ and the exchange constant $J$. Indeed $J$ is related to the critical temperature $T_c$. In the relationship between $J$ and $T_c$ one of the two two-spin operator averages takes place. These averages are:

$$p = \frac{1}{N} \sum_i \langle S_i^2 \rangle$$
$$q = \frac{1}{N} \sum_i \langle S_i \rangle^2.$$  

They are self-consistently calculated [3, 9] and the result generally depends on the temperature $T$. The temperature $T_c$ is determined by the condition:

$$kT_c = Jp_c$$  

where $p_c = p(T_c)$. $p_c$ is spin dependent; for $S = 1$, then $p_c = 0.790166$.

For $S = 1$ the spin operator $S_i = S_i^2$ is such as $S_i^3 = S_i$, $S_i^4 = S_i^2$ and consequently $r = p$ and $w = q$, while the inequalities $0 \leq q \leq p \leq 1$ hold.

In the paramagnetic phase ($T > T_c$) some cancellations happen: $q = 0$, $u = 0$, $v = 0$, $w = 0$ and therefore $s = p^2$. Then equation (2) becomes:

$$\frac{\delta c}{c_0} = - \frac{N^2 G^2 p(1-p)}{kT} \left( 1 - \frac{1}{2} \beta^2 J^2 p(1-p) \right)$$  

(S = 1 and $T \gg T_c$).

In contrast to the case of spins $S = 1/2$ the temperature variation of $\delta c/c_0$ is not a Curie law because of the temperature dependence of $p(T)$. However this simple law is asymptotically obtained at high temperatures $T \gg T_c$ when $\beta J$ tends towards $0$ and $p$ tends towards $2/3$.

In the spin-glass phase ($T \ll T_c$) equation (2) stays complicated. It must be emphasized that in this phase the SK solution of the model is not correct.

ii) For the Waller interaction the role of the coupling constant is played by a dimensionless coefficient $\gamma$ which measures the sensitivity of the exchange constants $J_{ij}$ to an elastic strain. We have found [3]:

$$\frac{\delta c}{c_0} = - \frac{N^2 \gamma^2 \beta}{2c_0} \left( (p^2 - q^2) + \frac{2 \beta^2 J^2}{B} [Ap^2 - Cq^2 + 4(u-w)pq] \right)$$  

where $A$ and $B$ are given by equations (3) and

$$C = 2(3v + s - 4u) - \beta^2 J^2 [(r-s)(3v+s-4u) + 2(u-w)^2].$$

3.1.2 Dynamical theory. — Glauber [12] and Suzuki and Kubo [13] have proposed a theory for the dynamics of magnetic systems. It has been applied to the case of spin-glasses where it allows the calculation of the elastic constant $c(\omega, T)$ [3, 14]. This is performed within the framework of the mean field theory corrected by the means of the reaction field [15, 16]. A phenomenological relaxation time $\tau(T)$, the same for every spin, has to be introduced.

The calculation can be achieved quite easily for Ising spins $S = 1/2$ because, in this case, the time derivatives of one-site and two-site averages ($\langle S_i \rangle$ and $\langle S_i S_j \rangle$) are expressed in terms of one-site and two-site averages only. This is no longer true for spins $S > 1/2$ because then higher order averages occur.
As a consequence of the assumption \( S = 1/2 \), the only possible coupling mechanism between spins and strains is the modulation of exchange interactions. Then our previous results [3] apply and the relative variation of the elastic constant turns out to be:

\[
\frac{\delta c}{c_0} = -\frac{N\gamma^2}{2c_0\beta} \left[ 1 - i\frac{\omega\tau}{2} F_1 \right]
\]

(9)

\( \omega/2\pi \) is the ultrasonic frequency.

\[
F_1 = \frac{1}{2} \left( \frac{T}{T_c} \right)^2 [1 + R_0 + i\omega\tau/2 - \sqrt{(1 + R_0 + i\omega\tau/2)^2 - 4(T_c/T)^2}]
\]

(10)

\( R_0 = R_0(T) \) is the reaction field; it is equal to \( (T_c/T)^2 \) in the paramagnetic phase.

In the static limit \( (\omega \rightarrow 0) \), equation (9) gives for \( T \rightarrow T_c \):

\[
\frac{\delta c}{c_0} = -\frac{N\gamma^2}{2c_0} \frac{T_c^2}{k} \frac{T}{T_c}
\]

(11)

At very high temperatures \( (T \gg T_c) \) the behaviour of equation (9) is:

\[
\frac{\delta c}{c_0} = -\frac{N\gamma^2}{2c_0} k \frac{T_c^2}{T} \frac{1}{1 + i\omega\tau/2}
\]

(12)

We recover a usual Debye relaxation equation.

At the critical temperature \( T_c \) the frequency dependence of \( \delta c/c_0 \) is unusual; for \( \omega\tau \ll 1 \) it is:

\[
\frac{\delta c}{c_0} = -\frac{N\gamma^2}{2c_0} kT_c \left[ 1 - \frac{\sqrt{\omega\tau}}{2} (1 + i) \right]
\]

(13)

3.1.3 Illustrated summary. — Figure 4 sums up the behaviour of the static elastic constant change according to different theories. This is done in reduced coordinates: \( \delta c(0, T)/\delta c(0, T_c) \) is given versus \( T/T_c \).

The solid line shows the prediction of the SK theory for \( S = 1 \) with a Van Vleck spin-phonon coupling, calculated according to equation (2).

The dashed line is obtained using the static limit \( (\omega \text{ tending towards } 0) \) of equation (9). It is explicitly given by equation (11) for \( T \gg T_c \). In this case the variation follows a Curie law.

Finally the dotted line reproduces the prediction of the SK theory for \( S = 1 \) with a Waller spin-phonon coupling as given by equation (8). This last contribution is shown as a reminder since the Van Vleck mechanism is expected to be dominant. This is confirmed by experiments (see Fig. 10).

3.2 Interpretation. — Our samples are both glasses and spin-glasses. As previously said, their elastic properties (and more precisely their complex elastic constant) must reflect this double nature and show the influence of the two-level systems (TLS) characteristic of the amorphous state and the influence of the spins.

The contribution of the TLS is well known [8]. In short, it is determined by the density of TLS and by the strength of their coupling with the elastic wave. Quantitatively only a few parameters are needed:

— two dimensionless parameters \( C_\sigma \) (\( \sigma = L \) or \( T \) for longitudinal or transverse) proportional to the TLS spectral density and to the square of the TLS-phonon coupling constants;
Fig. 4. — Magnetic contribution to the static acoustic susceptibility $\delta c/c_0$ normalized to 1 at its value at $T_c$, as a function of the reduced temperature $T/T_c$ calculated within three different expressions. Full line: SK theory with a Van Vleck spin-phonon coupling for $S = 1$. Dotted line: SK theory with a Waller spin-phonon coupling for $S = 1$. Dashed line: static limit of the dynamical theory (valid only for $S = 1/2$).

— a parameter $K_3$ occurring in the expression of the one-phonon relaxation rate;
— a dimensionless parameter $\mu$ which takes the deviation from the standard model into account (For more details, see [17]).

The values of $C_\sigma$, $K_3$ and $\mu$ are determined by experiments at temperatures far from the critical spin-glass temperature $T_c$ and at very high frequencies.

3.2.1 Nickel glass. — Figures 1 and 2 show the experimental data along with the calculated amorphous behaviours of the sound velocity $v_s$ and attenuation $\alpha$ as functions of temperature and frequency, obtained with our best determination of the unknown parameters $C_\sigma$, $K_3$ and $\mu$ given in table I. The differences between the theoretical curves and the data are ascribed to the spins.

These differences are reported in figures 5 and 6 after their transformation into the real and imaginary parts of $\delta c/c_0$ according to:

$$\text{Re} \left( \frac{\delta c}{c_0} \right) = 2 \frac{\delta v_s}{v_s}$$

$$\text{Im} \left( \frac{\delta c}{c_0} \right) = -\ln 10 \frac{v_s}{\omega} \alpha \quad (\alpha \text{ in dB/cm})$$

(14)
Fig. 5. — Difference between the experimental values of Re \((\delta c/c_0)\) (equal to twice \(\delta v/v_s\)) and the calculated values obtained from the TLS theory. The experimental data are those of figure 1 with the same symbols plus one set of data at 926 MHz. From the bottom to the top: crosses: 31 MHz; stars: 92 MHz; open circles: 330 MHz; crosses: 926 MHz; plusses: 1556 MHz.

with

\[ c_0 = \rho v_s^2 \quad (\rho \text{ is the density}). \]

The real part of \(\delta c/c_0\) is negative (the interaction with spins slows down the acoustic wave). Its variation with \(T\) presents a minimum while at a given temperature its magnitude is a decreasing function of frequency. The imaginary part of \(\delta c/c_0\) is positive and as a function of \(T\) it presents a maximum.

We may bring out the Ariadne's clew of our approach as follows. We first assume that the dynamical change of the elastic constant is the product of a static change by a dynamical factor:

\[ \frac{\delta c(\omega, T)}{c_0} = \frac{\delta c(0, T)}{c_0} D(\omega, T). \]

The calculation of \(D(\omega, T)\) can be achieved only for \(S = 1/2\); it is equal to the function between brackets in equation (9) multiplied by \((\beta \bar{J})^2\). Under this hypothesis we are able to extract the static behaviour \(\delta c(0, T)/c_0\). In a second step, we forget the procedure and we evaluate \(\delta c(0, T)/c_0\) for different models: \(S = 1/2\) or \(S = 1\); Van Vleck or Waller coupling mechanisms.
Therefore the dynamical behaviour is first evaluated with the help of charts drawn from equations (9) and (10). This is shown in figures 7 and 8 for two temperatures corresponding to two characteristic variations. At $T = T_c$ [18] (Fig. 7) there is clear evidence for an $\omega^{1/2}$ variation rather than for a Debye relaxation law. On the contrary at $T > T_c$ (Fig. 8) the Debye relaxation and the dynamical theory give neighbouring predictions. In this way we could obtain the static elastic constant change $\delta c(0, T)/c_0$ and the relaxation time $\tau(T)$ for every temperature.

The relaxation time of the Ni glass has values in the nanosecond range and presents no anomaly at $T_c$. This is shown in figure 9.

The absolute value of the magnetic contribution to the static elastic constant (see Fig. 10) shows a maximum near $T_c$. In this figure our experimental data with the curve giving the best fit (static SK model with a Van Vleck mechanism for $S = 1$) are given. To our satisfaction, the value of the coupling constant which we can deduce ($G = 6.5 \times 10^{-22} \text{ J} \approx 33 \text{ cm}^{-1}$) is in agreement with what is known for Ni$^{2+}$ [5].

3.2.2 Cobalt glass. — The results concerning the cobalt glass have been extensively reported elsewhere [2, 3]. Only few of them are recalled.

Figures 3 and 9 present some data on the sound velocity and relaxation time. The aim of
Fig. 7. — Simultaneous fit of the magnetic contribution to Re (δc/ε₀) (open circles) (the sign of which has been changed) and Im (δc/ε₀) (stars) to the theoretical curves deduced from equations (12) (Debye relaxation, dotted lines) and from equation (9) (dynamical theory, solid lines). The experimental data are obtained for different frequencies at a given temperature (here T = Tc = 1.5 K).

3.2.3 Manganese glass. — In figure 3 results obtained on a manganese glass are also shown. They were published with an interpretation which neglected the spin-glass effect [4]. After we have performed our experiments on cobalt and nickel glasses, we revisited our first interpretation, slightly changing the TLS-parameter K₃. We obtain the curve drawn in the figure. The data points clearly show that a negative contribution is superposed on the glassy features. This contribution is ascribed to the spin-glass transition. The fact that all these magnetic fluorophosphate glasses behave similarly is thus brought into evidence.

Even with this new point of view, the anomalous features quoted in [4] remain true: in this magnetic glass the spectral density of the TLS is very small while their coupling to phonons is rather strong. This anomaly is not explained.

3.3 DISCUSSION. — The thesis we advocate in this article is clearly demonstrated in figure 3. The sound velocity data from a non-magnetic fluorophosphate glass shows the typical behaviour of a standard insulating glass. On the contrary the low-frequency (ωτ ≪ 1)
measured sound velocities of three magnetic glasses all present the same feature: superposed on the glassy behaviour there is a negative magnetic contribution.

In the cases of nickel and cobalt glasses where our data are more numerous and accurate we are able to extract from the attenuation and velocity sound measurements the temperature dependence of the relaxation time $\tau(T)$ and of the static elastic constant $\delta C(\omega = 0, T)/C_0$.

For both glasses the relaxation time fits reasonably well an Arrhenius law $\tau = \tau_0 \exp(T_0/T)$ as seen in figure 9. Table II gives the values of $\tau_0$ and $T_0$. In the two cases, the temperature which measures the barrier height is $T_0 \approx 2T_c$, while $\tau_0$ falls in the $10^{-10}-10^{-11}$ s range.

Since our experiments provide the value of the spin-phonon coupling constant $G$ we can calculate the spin-lattice relaxation time $T_1$ due to a direct (one-phonon) process of an individual spin. We notice that this point of view is coherent with the main assumption of Glauber [12] about the relaxation time $\tau$ but implies an off-diagonal spin-phonon coupling Hamiltonian. Assuming a typical splitting of the spin levels equal to $J$, the relaxation rate is:

$$\Gamma_1(J, T) = \frac{G^2}{8 \pi \rho v^5 \hbar^4} J^3 \coth(\beta J/2).$$

This gives $T_1 = \Gamma_1^{-1} \approx 10^{-4}$ s in the vicinity of $T_c$. This value which must be compared to the experimental datum $\tau \approx 4 \times 10^{-10}$ s at $T_c$, rules out such a relaxation as the dominant process.
Fig. 9. Variation of the relaxation time $\tau(T)$ deduced from the acoustic experiments using the dynamical theory (see text). It is sketched on an Arrhenius plot (logarithm of the inverse of $\tau$ as a function of the inverse of the temperature). The data concerning the Ni glass are represented by the open circles. The stars represent the data obtained previously [3] for the Co glass. The full and dashed lines are the best Arrhenius fits whose characteristic parameters are given in table II. Vertical arrows show the two critical temperatures $T_c$(Ni) and $T_c$(Co).

Table II. Magnetic parameters for the three magnetic glasses the exact composition of which is given in table I: effective spin, spin-freezing temperature $T_c$, number of magnetic ions per unit volume $N$, $\tau_0$ and $E_0$ parameters of the Arrhenius fit of the relaxation time deduced from acoustic experiments.

<table>
<thead>
<tr>
<th></th>
<th>Mn</th>
<th>Ni</th>
<th>Co</th>
</tr>
</thead>
<tbody>
<tr>
<td>$S$</td>
<td>5/2</td>
<td>1</td>
<td>1/2</td>
</tr>
<tr>
<td>$T_c$ (K)</td>
<td>3</td>
<td>1.5</td>
<td>1.8</td>
</tr>
<tr>
<td>$N$ (m$^{-3}$)</td>
<td>$1.43 \times 10^{28}$</td>
<td>$1.06 \times 10^{28}$</td>
<td>$1.02 \times 10^{28}$</td>
</tr>
<tr>
<td>$\tau_0$ (s)</td>
<td>—</td>
<td>$5.4 \times 10^{-11}$</td>
<td>$3 \times 10^{-10}$</td>
</tr>
<tr>
<td>$E_0/k$ (K)</td>
<td>—</td>
<td>3.5</td>
<td>3.6</td>
</tr>
</tbody>
</table>
Fig. 10. — Magnetic part of the opposite of the static elastic constant in the spin-glass 
\((\text{NiF}_2)_3(\text{BaF}_2)_2(\text{NaPO}_3)_3\) as a function of temperature. The open circles are the experimental data 
deduced from acoustic experiments. Solid line corresponds to the SK theory using a Van Vleck spin-
phonon coupling for spins \(S = 1\).

The observed relaxation time is so short that a specific process must probably be considered.

However the most surprising result is the apparent adequacy of the mean field theory to 
experimental data obtained on materials with short-range magnetic, probably non-Gaussian 
distributed, interactions. Another astonishing result is the non-criticality of the unique 
relaxation time \(\tau\) at \(T_c\), especially since measurements of the magnetic susceptibility give 
evidence for critical characteristic times, with exponents which are not those of the mean field 
theory.

In order to go further in the understanding of these features (and more generally of the SG 
state) more theoretical and experimental efforts are needed on : Glauber's dynamics of SG 
for \(S > 1/2\), non mean field dynamics, acoustic experiments at low frequencies and the 
relationship between Glauber's point of view with a unique relaxation time and a distribution 
of Debye relaxations.

4. Conclusion.

We have reported on acoustic propagation experiments in several insulating amorphous spin-
glasses. Our data on a Ni glass confirm previous results obtained in a Co glass. The frequency
dependent part of the magnetic contribution is well explained within the framework of a dynamical theory. In particular an $\omega^{1/2}$ law is observed at $T = T_c$. The static limit is accounted for by a SK theory using a spin-phonon coupling by a Van Vleck mechanism for the Ni glass ($S = 1$) while a Waller mechanism was used for the Co glass ($S = 1/2$). Comparison between four glasses emphasizes the general validity of the approaches used in this article.

References

[18] The value $T_c = 1.5$ K given by our ultrasonic experiments is somewhat different of $T_c \approx 2.5$ K as obtained from magnetic susceptibility measurements performed on another sample of the same nominal composition (J.-P. Renard : private communication).