Evidence for a two-component tunnelling mechanism in the multicomponent glasses at low temperatures

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Abstract – The thermal and dielectric anomalies of window-type glasses at low temperatures \((T < 1 \text{K})\) are somewhat successfully explained by the two-level systems (2LS) tunneling model (TM). However, the magnetic effects discovered in the multisilicate glasses in recent times, and also some unexplained older data in zero magnetic field for the mixed \((\text{SiO}_2)_{1-x} (\text{K}_2\text{O})_x\) and \((\text{SiO}_2)_{1-x} (\text{Na}_2\text{O})_x\) glasses, indicate the need for a suitable generalization of this standard model. We present evidence that also for the mixed glasses in the absence of a field the right extension of the 2LS TM is provided by the multilevel (anomalous) tunneling systems approach proposed by one of us to explain the magnetic effects. It appears that new 2LS develop via dilution near the hull of the \text{SiO}_2-percolating clusters in the mixed glasses.

Tunneling systems (TS) are useful for many theoretical investigations in condensed-matter physics: from laser-matter interaction, to amorphous solids and disordered crystals, to hydrogen storage in metals [1]. For their simplicity, only the two-level systems (2LS) are usually employed instead of the more realistic multilevel systems; in this letter a situation will be discussed where the use of a number of states greater than two is essential in order to make progress. Moreover, new insight will be given on the role of percolation and fractal theory in the context of the tunneling model (TM) for the multicomponent glasses.

Glasses at low temperatures are interesting for their rather universal physical properties attributed to the low-energy excitations characterising all kinds of amorphous solids. The 2LS TM has been somewhat successful in the semi-quantitative explanation of a variety of interesting thermal, dielectric and acoustic anomalies of structural glasses at temperatures \(T < 1 \text{K}\) [2–5]. This model assumes that the important degrees of freedom derive from “particles” tunneling between the wells of a generic double-welled potential with broad well-asymmetry and barrier-height distribution in the solid. However, no clear identification of the nature of these particles has been achieved so far, after almost 40 years of the model’s proposition. A more precise identification remains therefore highly desirable. The basic physics of cold glasses is important also because of its link with the mechanism of the glass freezing transition [6,7]. From the experimental and theoretical points of view, beside the linear in \(T\) behavior of the heat capacity \(C_p\), it is believed that the linear in \(\pm \ln T\) behavior (V-shaped curve) of the real part of the frequency-dependent dielectric constant \(\epsilon'(\omega)\) represents a cogent characterization of the glassy state at low temperatures. Here we address the question of how satisfactory our understanding of the experimental situation is and whether the 2LS approach is indeed the full physical picture.

Figure 1 (inset) shows the behavior of the \(T\)-dependent part of \(\epsilon'\), \(\Delta \epsilon'/\epsilon' = [\epsilon'(T) - \epsilon'(T_0)]/\epsilon'(T_0)\), (where \(T_0(\omega)\) is a characteristic minimum) for pure vitreous SiO\(_2\), free of contaminants: a paradigm glassy solid. It can be seen that linear regimes in \(-\ln T\) for \(T < T_0\) and \(+\ln T\) for \(T > T_0\) are observed, and roughly with \(\ln T\) slopes \(S_- = -2S\) and \(S_+ = S > 0\), respectively, or in a \(-2:1\) ratio as one finds in the literature. According to the 2LS TM, in fact, we have the following standard expressions for the resonant (RES) and relaxational (REL) tunneling contributions to

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Fig. 1: Dielectric signature of pure $a$-SiO$_2$ (inset) and AlBaSiO (main) glasses. SiO$_2$ data [9], fitted with eq. (1), display a $-2:1$ 2LS TM behavior. AlBaSiO data [10,11] display rather a $-1:1$ behavior, yet could be fitted with eq. (1) (dashed line) [12] with a large $\Delta_{0\text{min}} = 12.2 \text{mK}$ 2LS tunneling parameter. We have fitted all data with a more realistic $\Delta_{0\text{min}} = 3.9 \text{mK}$ and best-fit parameters from table 1 using eqs. (1) and (9) (driving frequency $\omega = 1 \text{kHz}$). KFS stands here for ref. [12].

the dielectric constant (see, e.g., [1–5,8]):

$$\frac{\Delta'\varepsilon'}{\varepsilon'}_{2\text{LS}} = \frac{\Delta'\varepsilon'}{\varepsilon'}_{2\text{RES}} + \frac{\Delta'\varepsilon'}{\varepsilon'}_{2\text{REL}},$$

$$\frac{\Delta'\varepsilon'}{\varepsilon'}_{2\text{RES}} = \frac{2P_0^2}{3\omega \varepsilon_r} \int_{z_{\text{min}}}^{z_{\text{max}}} dz \left( \frac{\Delta_{0\text{min}}}{2k_B T z} \right)^2 \tanh z,$$

$$\frac{\Delta'\varepsilon'}{\varepsilon'}_{2\text{REL}} = \frac{P_0^2}{3\omega \varepsilon_r} \int_{z_{\text{min}}}^{z_{\text{max}}} dz \int_{\tau_{\text{min}}}^{\tau_{\text{max}}} \frac{d\tau}{\tau}$$

$$\times \left( 1 - \frac{\tau_{\text{min}}}{\tau} \cosh^{-2}(z) \right),$$

where we neglect, for low $\omega$, the frequency dependence in the RES part. Here $z_{\text{min,max}} = \Delta_{0\text{min,max}}/2k_B T$ and $\tau$ is the phenomenological 2LS relaxation time given by, with $E = 2k_B T z$,

$$\tau(E)^{-1} = E\Delta^2_0/\gamma \tanh \left( \frac{E}{2k_B T} \right).$$

Also, $\Delta_0$ is the tunneling parameter of a 2LS having energy gap $E$, $\Delta_{0\text{min}}$ and $\Delta_{0\text{max}}$ are its phenomenological bounds, $\gamma$ is a further (elastic) material parameter of the solid and $\tau_{\text{min}}^{-1} = E^3/\gamma \tanh \left( \frac{E}{2k_B T} \right)$, $\tau_{\text{max}}^{-1} = E\Delta^2_{0\text{min}}/\gamma \tanh \left( \frac{E}{2k_B T} \right). P$ is the probability per unit volume and energy that a 2LS occurs in the solid and $p_0^2$ is the average square 2LS electric-dipole moment.

Indeed, from expressions (1) we see that (after standard manipulation of the integrals): 1) the RES contribution has the leading behavior

$$\frac{\Delta'\varepsilon'}{\varepsilon'}_{2\text{RES}} \simeq \begin{cases} -\frac{2P_0^2}{3\omega \varepsilon_r} \ln \left( \frac{2k_B T}{\Delta_{0\text{max}}} \right), & \text{if } T < \frac{\Delta_{0\text{max}}}{2k_B}, \\ 0, & \text{if } T > \frac{\Delta_{0\text{max}}}{2k_B}. \end{cases}$$

Thus, the sum of the two contributions has a V-shaped form, in a semilogarithmic plot, with the minimum occurring at a $T_0$ roughly given by the condition $\omega_{\tau_{\text{min}}}(k_B T) \approx 1$, or $\omega_{T_0}(\omega) \approx \left( \frac{3}{2} \right)^{1/2} \varepsilon_r$. Here is the bulk of the solid’s dielectric constant and we see that a $-2:1$ characteristic behavior is justified by the TM with the slope for $T > T_0$ given by $S = \frac{P_0^2}{3\omega \varepsilon_r}$.

This behavior is observed in pure $a$-SiO$_2$ [9] (with the parameters of table 1, $x = 0$, from our own best fit to eq. (1)). However in most multicomponent glasses (structurally made up of good glass formers and of good crystal formers) it is rather a V-shaped curve with a (roughly) $-1:1$ slope ratio that is often observed. Figure 1 (main panel) shows this phenomenon for the multisliscate glass Al$_2$O$_3$-BaO-SiO$_2$ (in short, AlBaSiO), which has been extensively investigated in recent times due to its unexpected magnetic field response [10–14]. Also, fig. 2 shows the behavior of the dielectric constant $\varepsilon$ vs. $T$ for the glasses of composition $\text{(SiO}_2\text{)}_{1-x} \cdot \text{(K}_2\text{O)}_x$ containing a molar concentration $x$ of alkali oxide [15]. It is seen that a $S_-/S_+$ slope ratio closer to $-1:1$ is observed, with the slope definitely changing with $x$, and faster for $T > T_0$. These data, thus far unexplained by the 2LS TM to our knowledge, call for an extension of the accepted TM and we show in this letter that a simple explanation can be given in terms of the very same additional (anomalous) tunneling systems (ATS) that have been advocated by one of us to explain the magnetic response of AlBaSiO and other multicomponent glasses [13,14]. In view of the interest for these materials in low-$T$ thermometry, and on fundamental grounds, such explanation appears overdue to us. Moreover, “additional” TS (beside the standard 2LS) were already called for in [15] and earlier papers, though never identified as precisely as in our work.

In a rather general fashion, the TS can be thought of as arising from the shape of the theoretical energy-landscape $\mathcal{E}(\{r_i\})$ of a glass as $T$ is lowered well below the glass freezing transition $T_f$. Many local and global minima develop in $\mathcal{E}(\{r_i\})$ as $T \to 0$, the lowest-energy minima of interest being made up of multiple $n_w = 2, 3, \ldots$ local wells separated by shallow barriers. These local multwelled potentials are our TS and it seems reasonable that the $n_w = 2$-wellged potentials should be ubiquitous.
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Table 1: Extracted parameters for the glasses; K-Si stands for the \((\text{SiO}_2)_{1-x}(\text{K}_2\text{O})_x\) glasses. In all of the best fits we have employed the values \(\Delta_{0\text{min}} = 3.9\ \text{mK}\) and \(\Delta_{0\text{max}} = 10\ \text{K}\) extracted from fitting the pure \text{SiO}_2 data of fig. 1 (inset).

| Glass type | \(x\) | \(A_{2L5}\times10^{-5}\) | \(\gamma\times10^{-8}\) | \(A_{ATS}\times10^{-5}\) | \(D_{\text{min}}\) | \(\Gamma\) |
|------------|-------|--------------------------|-----------------|------------------|-----------------|----------|
| \text{SiO}_2 | 0.00 | 47.2 | 5.30 | – | – | – |
| AlBaSiO   | –    | 116.2 | 13.40 | 264.7 | 0.65 | 69.73 |
| K-Si 0.05 | 104.1 | 1.33 | 75.5 | 0.87 | 3.55 |
| K-Si 0.08 | 146.5 | 1.23 | 130.0 | 0.87 | 3.97 |
| K-Si 0.10 | 158.5 | 1.15 | 160.0 | 0.87 | 5.08 |
| K-Si 0.20 | 239.5 | 0.82 | 281.9 | 0.87 | 6.44 |

Fig. 2: Dielectric signature of mixed \((\text{SiO}_2)_{1-x}(\text{K}_2\text{O})_x\) glasses as functions of \(T\) and \(x\) [15]. Fitting parameters from table 1 using eqs. (1) and (9) from our theory (driving frequency \(\omega = 10\ \text{kHz}\)).

We stress that the term “devitrification” means not necessarily microcrystal formation, though microcrystals have been observed in the multicomponent glasses [17–19]. Here we mean by this term the tendency to form regions of enhanced atomic-arrangement regularity as is to be expected when introducing good crystal-forming species (BaO, CaO, K\(_2\)O, Na\(_2\)O etc.) within a good glass-former’s (SiO\(_2\), B\(_2\)O\(_3\), etc.) environment. Simulations and experiments in the multisilicates have shown that the NM-species partly destroy the networking capacity of the NF-ions and form their own pockets and channels inside the NF-network [20]. Hence, \(n_{w} > 2\) multiwelled TS can be imagined to arise inside these NM-pockets and -channels and should be described by some new energy-parameters’ distribution when some degree of “devitrification” occurs, leading to entirely new physics. Simulations [21,22] have shown the occurrence of multiwelled local tunneling potentials as likely.

One of the present Authors has proposed that precisely this situation occurs inside the magnetic-sensitive multicomponent glasses [13,14], and in this letter we show how this proposal appears to explain the zero-magnetic-field dielectric data of figs. 1–3 as well. We remind the reader that our proposal consists in using, beside the standard
2LS each described by the reduced Hamiltonian

$$H_{2LS} = -\frac{1}{2} \left( \frac{\Delta}{\Delta_0 - \Delta} \right)$$

and double-well potential parameters distributed according to

$$\mathcal{P}_{2LS}(\Delta, \Delta_0) = \frac{\bar{P}}{\Delta_0},$$

also a collection of ATS with (for simplicity) \( n_w = 3 \), each described by the Hamiltonian

$$H_{ATS} = \begin{pmatrix} E_1 & D_0 & D_0 \\ D_0 & E_2 & D_0 \\ D_0 & D_0 & E_3 \end{pmatrix}$$

and with triple-well potential parameters \( E_1, E_2, E_3 \) (well asymmetries) and \( D_0 \) (maximum well transparency) distributed according to

$$\mathcal{P}_{ATS}(E_1, E_2, E_3, D_0) = \frac{P^*}{D_0(E_1^2 + E_2^2 + E_3^2)}$$

(\( P^* \) being a new material parameter). The above is the simplest modification of the standard TM distribution, eq. (6), allowing, with appropriate cutoffs, for some amount of devitrification and explaining the magnetothermal anomalies of the multicomponent glasses [13]. As such it does not contain, however, information on the spatial extent of the enhanced atomic regularity regions. Using this theory for \( D_0 \gg \sqrt{E_1^2 + E_2^2 + E_3^2} \equiv D \) as described in [13,14], we arrive at the expression for the RES and REL contributions to the dielectric anomaly from the advocated ATS:

$$\frac{\Delta \epsilon'}{\epsilon'}_{ATS} = \frac{\Delta \epsilon'}{\epsilon'}_{ARES} + \frac{\Delta \epsilon'}{\epsilon'}_{AREL},$$

$$\frac{\Delta \epsilon'}{\epsilon'}_{ARES} = \frac{3\pi P^* p_1^2}{2 \epsilon_0 \epsilon_r D_{min}} \int_1^\infty \frac{dy}{y^2 \tanh \left( \frac{D_{min}}{2k_B T} y \right)},$$

$$\frac{\Delta \epsilon'}{\epsilon'}_{AREL} = \frac{\pi P^* p_1^2}{2 \epsilon_0 \epsilon_r D_{min}} \left( \frac{D_{min}}{2k_B T} \right)$$

$$\times \int_1^\infty \frac{dy}{y} \cosh^{-2} \left( \frac{D_{min}}{2k_B T} y \right) \frac{1}{1 + \omega^2 \tau_{ATS}^{-1}}.$$  

Here we have again neglected, for low-\( \omega \), the frequency dependence in the RES part, we put \( y = D/D_{min} \) and \( \tau_{ATS} \) is the largest phenomenological ATS relaxation time given by

$$\tau_{ATS}^{-1} = D^5 / \Gamma \tanh \left( \frac{D}{2k_B T} \right).$$

Moreover, \( D_{min} \) is now the lower bound for the energy gap of a multilevel ATS, \( \Gamma \) is the appropriate ATS elastic constant and \( P^* \) is the (slightly renormalised) probability per unit volume that an ATS occurs in the NM pockets and channels, with \( \bar{p}^2 \) the average square ATS dipole moment. This description is linked to the distribution function of eq. (8) [13,14] for these ATS favoring near-degenerate energy gaps bound from below, however, by a \( D_{min} > 0 \). In turn, this produces an overall density of states

$$g(E) = g_{2LS} + g_{ATS}(E) \approx 2\bar{P} + \frac{2\pi \bar{P}^*}{E} (E - D_{min}),$$

that is roughly of the form advocated in [15] and by other contemporary Authors (e.g., [23,24]) to explain anomalies in the glasses not accounted for by the standard 2LS TM. Here, we associate these phenomenological additional degrees of freedom with ATS arising through partial “devitrification” within the NM regions.

Manipulation of the expressions in (9) shows that: 1) the RES contribution from the ATS has the leading behavior (for \( T < D_{min}/2k_B \), \( \epsilon'_{ARES} \) is a constant)

$$\left. \frac{\Delta \epsilon'}{\epsilon'} \right|_{ARES} \approx \begin{cases} 0, & \text{if } T < \frac{D_{min}}{2k_B} \\ \frac{\pi \bar{P}^* p_1^2}{6 \epsilon_0 \epsilon_r k_B T} \ln \left( \frac{2k_B T}{D_{min}} \right), & \text{if } T > \frac{D_{min}}{2k_B} \end{cases},$$

2) the REL contribution is, instead, characterised by the leading form

$$\left. \frac{\Delta \epsilon'}{\epsilon'} \right|_{AREL} \approx \begin{cases} 0, & \text{if } \omega \tau_{ATS} \gg 1, \\ \frac{\pi \bar{P}^* p_1^2}{\epsilon_0 \epsilon_r k_B T} \ln \left( \frac{k_B T}{D_{min}} \right), & \text{if } \omega \tau_{ATS} \ll 1. \end{cases}$$

Thus, the V-shaped semilogarithmic curve is somewhat lost. However adding the 2LS (eq. (1)) and ATS (eq. (9)) contributions together one does recover a V-shaped curve with a slope \( S_\perp - 2S \) basically unchanged for \( T < T_0 \) and an augmented slope \( S_\parallel = S + S_{ATS} \) for \( T > T_0 \) with \( S_{ATS} = \pi P^* p_1^2 / 6 \epsilon_0 \epsilon_r k_B T \) that for \( T < D_{min}/k_B \) may approach 2S and thus (qualitatively) a -1:1 slope ratio.

We have fitted expressions (1) and (9) to the data for AlBaSiO in fig. 1 (main panel) and to the x-dependent data for SiO\( \text{2}(1-x)K_2O \) in figs. 2 and 3, obtaining in all cases very good agreement between theory and experiments. Figure 3 shows the fit of our theory to the frequency-dependent data for \( x = 0.2 \). In all these best fits we have kept the value of \( \Delta_{min} = 3.9 \text{mK} \) fixed, as obtained from our pure-SiO\( \text{2} \) fit, and the value of \( D_{min} \) also independent of \( x \) and \( \omega \). The idea is that these parameters are rather local ones and should not be influenced by NF/NM dilution. Table 1 reports all the (2LS and ATS) parameters used for our best fits, where we have defined the prefactors \( A_{ATS} = \pi \bar{P}^* p_1^2 / \epsilon_0 \epsilon_r D_{min} \) and \( A_{2LS} = \bar{P}_{D0} / \epsilon_0 \epsilon_r \). For AlBaSiO the parameters obtained differ somewhat from the values \( A_{ATS} = 1.42 \times 10^{-3} \) and \( D_{min} = 0.03 \text{K} \) used in [14] (where however a best fit for the magnetic-field (B) dependence of \( \epsilon' \) at very low

\( ^2 \text{TS-TS interactions can be neglected above } T \sim 100 \text{mK.} \)
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The scaling with $x$ of the prefactor $A_{ATS}$ (hence, ultimately, of the parameter $P^*$ in eq. (8)) is the main result of this paper. It shows that there are extra degrees of freedom of the tunneling type already advocated for the magnetic-field effect in the multicomponent glasses that give a contribution scaling like the dilution $x$ of the NM-species in the system $(\text{SiO}_2)_{1-x}(\text{K}_2\text{O})_x$. This gives a definite footing for the existence of our ATS in these amorphous solids. Our theory, with parameters from table 1, and new prefactors, can also account for the $T$- and $x$-dependence of the heat capacity $C_p(T,x)$ (data from [15]) confirming the above fractal picture. The two-component tunneling mechanism we advocate has been able to explain semi-quantitatively, and with roughly the same phenomenological parameters, the experimental data for $C_p(T,B)$ [13], $\epsilon'(T,B)$ [14] and now also for $C_p(T,x)$ and $\epsilon'(T,x)$ in the multicomponent silicate glasses. We therefore believe it is the correct simplest extension of the 2LS TM for these multiphase glasses.

The use of multiwelled tunneling potentials is not new in this field in order to improve the energy landscape description for the cold glasses. Three-level TS similar to ours —though with a much higher-energy third well and a broad distribution of tunneling parameters as in eq. (6)— have been advocated to understand the out-of-equilibrium response in strong dc electric fields [25]. Also, the existence of rugged-barrier (hence, effectively, multiwelled) standard 2LS has been advocated in order to explain the magnetocapacitance effect in the $\text{SiO}_2+\text{C}_y\text{H}_2$ glasses [26]. Only our ATS, however, obey the new distribution, eq. (8), resulting in an enhancement of the Aharonov-Bohm orbital coupling to the magnetic field.

As will be shown elsewhere, the use of these ATS improves the agreement between theory and experiments, especially for $T>T_0$, also for the dielectric loss $\tan \delta = \epsilon''/\epsilon'$ ( $\epsilon''$ being the imaginary part of the frequency-dependent dielectric constant). Though we are unaware of data for the dielectric loss for the mixed glasses, for $\text{AlBaSiO}$ (and with the parameters of table 1) only a qualitative fit to the available data [11] can be achieved for $\tan \delta$ within our theory too. In fact, the TM is unable to provide a good fit simultaneously for both $\epsilon'$ and $\tan \delta$ within the relaxation time approximation which we have used.

In summary, we have shown that there is direct evidence in zero magnetic field already for the multiwelled ATS advocated to explain the magnetic-field effect in the multicomponent glasses. The relevance of multiwelled TS in the multicomponent glasses is a new and unexpected finding in this field of research. Our work predicts that the magnetic response of the alkali-silicate glasses should be important and scale like the molar alkali concentration $x$. At the same time the $-1:1$ slope ratio problem of the standard TM has been given a simple explanation in terms of our two-component tunneling model. For AlBaSiO, using the value of $\tilde{P}^*$ extracted from $C_p$ [13] and that

![Fig. 4: The 2LS and ATS prefactor parameters ($\times 10^5$) for all glasses (from table 1) as a function of $x$. Our data fit well with our theoretical expectations (full lines).](image-url)
of $A_{ATS}$ given in table 1, we estimate the value of the electric dipole of the effective anomalous tunneling entity: $p_{eff} \equiv \sqrt{p_1^2} \approx 0.4$ D. This small value confirms [13,14] that this new physics must come from the coherent tunneling of a small ionic cluster (the very same origin of the large values of $D_{min}$ and [13,14] of $D_{min}, D_{max}$). Details of our present analysis will be published elsewhere, together with our study of the mixed glasses’ heat capacity data. The latter allows us to estimate the effective ATS electric dipole for the mixed glasses: $p_{eff} \approx 0.05$ D, independent of $x$, corroborating the idea of a small coherent tunneling cluster being represented by our fictitious tunneling particle. Indeed, the dipole moment of a cluster of $N$ coherently tunneling particles $p_{eff} = \sum_{i=1}^{N} p$, can become much smaller than the elementary atomic dipole $EO = 2.54$ D ($e$ is the elementary charge and $a_0$ the Bohr radius). Conversely, the tunneling parameters $D$ and especially $D_0$ become roughly the arithmetical sums of those of the bare coherently tunneling particles, $D \sim ND_0$ and $D_0 \sim N D_0$, respectively. Therefore, since $N$ can attain values as large as 200 [7] (independently of the solid’s composition) in some models, this leads to values of $D_i$ and $D_0$ comparable to those characteristic of the 2LS TM. Despite the (only apparently) large values of the cluster’s $D_{min}$ (table 1), the temperature dependence of the dielectric constant is sufficiently strong in our approach as long as $D \leq 2k_BT$, as guaranteed by the distribution equation (8).

The present work suggests the way new experiments on mixed glasses could help in deciding whether our own two-component tunneling mechanism [13,14] or the more exotic nuclear quadrupole-moment approach [27] is the correct explanation for the magnetic-field effects in the cold glasses. Amorphous solids of composition (SiO$_2$)$_{1-x}$(MO)$_x$ should be investigated for different concentrations $x$ of NM-species at ultralow temperatures and in the presence of a magnetic field. If $M$ is a divalent metal (ideally Ca, but also Mg, Sr and Ba) with no or a tiny percentage of stable isotopes possessing a nonzero nuclear quadrupole moment, then a magnetic response scaling like $x$ would single out the present approach. The system (SiO$_2$)$_{1-x}$(CaO)$_x$ (with a small percent of PbO favoring nanocrystal formation) appears to be the ideal candidate. We remark, incidentally, that in [26] the amorphous system SiO$_{2+x}CaH_x$, devoid of nuclear quadrupole moments, displayed magnetic effects quantitatively similar to those found in glasses with nuclei carrying such moments at ultralow temperatures.

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