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# Dielectric susceptibility and heat capacity of ultra-cold glasses in magnetic fields

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## Abstract

Recent experiments demonstrated unexpected, even intriguing properties of certain glassy materials in magnetic fields at low temperatures. We have studied the magnetic field dependence of the static dielectric susceptibility and the heat capacity of glasses at low temperatures. We present a theory in which we consider the coupling of the tunnelling motion to nuclear quadrupoles in order to evaluate the static dielectric susceptibility. In the limit of weak magnetic field we find the resonant part of the susceptibility increasing like  $B^2$  while for large magnetic field it behaves as  $1/B$ . In the same manner we consider the coupling of the tunnelling motion to nuclear quadrupoles and angular momentum of tunnelling particles in order to find the heat capacity. Our results show the Schottky peak for the angular momentum part, and  $B^2$  dependence for the nuclear quadrupole part of the heat capacity, respectively. We discuss whether or not this approach can provide a suitable explanation for such magnetic properties.

## 1. Introduction

At very low temperatures, glasses and other amorphous systems show similar thermal, acoustic and dielectric properties [1], which are in turn very different from those of crystalline solids. Below 1 K, the specific heat  $C_v$  of dielectric glasses is much larger and the thermal conductivity  $\kappa$  orders of magnitude lower than the corresponding values found in their crystalline counterparts.  $C_v$  depends approximately linearly and  $\kappa$  almost quadratically on temperature [2], respectively. This is in clear contrast to the cubic dependence observed in crystals for both properties, well understood in terms of Debye's theory of lattice vibrations. Above 1 K,  $C_v$  still deviates strongly from the expected cubic dependence, exhibiting a hump in  $C_v/T^3$  which is directly related to the so-called boson peak observed by neutron or Raman vibrational spectroscopies [3]. To explain these, it was considered that atoms, or groups of

atoms, are tunnelling between two equilibrium positions, the two minima of a double-well potential. The model is known as the two-level system (TLS) [4, 5]. In the standard TLS model, these tunnelling excitations are considered as independent, and some specific assumptions are made regarding the parameters that characterize them (for a review see for example [1]).

In recent years an intriguing magnetic field dependence of the dielectric and coherent properties of some insulating glasses was reported. In 1998 Strehlow *et al* observed a sharp kink at  $T = 5.84$  mK in the dielectric constant of the multi-component glass BaO–Al<sub>2</sub>O<sub>3</sub>–SiO<sub>2</sub> [6] measured in very weak magnetic field, of the order of  $10 \mu\text{T}$ . The effect was several orders of magnitude larger than what is expected, considering the absence of magnetic impurities in this insulator. Later studies carried on in magnetic field ranging up to 25 T and temperatures well below 100 mK revealed for several materials a complex and strong dependence of the dielectric response on the external magnetic field, on the applied voltage and on temperature [6–10]. Even more surprising phenomena were observed in the spontaneous polarization echo experiments on BaO–Al<sub>2</sub>O<sub>3</sub>–SiO<sub>2</sub> [11]. The increase of the echo amplitude by a factor of four was reported when varying the magnetic field in the range of 0–200 mT. Similar results were later reported in the case of amorphous mixed crystal KBr<sub>1-x</sub>CN<sub>x</sub> [12]. To date a unified theory does not exist while some contradictions are present in the recent works. We have clear evidence that such intriguing magnetic properties are connected with the low-energy tunnelling excitations present in almost all amorphous solids. These tunnelling states are known to be proved very well by the echo experiments [14]. On the other hand these excitations have been reported as showing up at very low temperatures (usually  $T < 100$  mK), where the TLSs are responsible for the thermal and dynamical properties of glasses [12, 13].

Several generalizations of the standard TLS model have been reported after the anomalous behaviour of glasses in a magnetic field. The main question for such models is how a TLS should interact with the magnetic field, it being not clear how the tunnelling entity would acquire a finite magnetic moment. According to the proposed solutions, the models can be divided into ‘orbital’ and ‘spin’ models.

In the orbital models, the tunnelling entities are not simple two-state systems, but perform some kind of circular motion. Due to the presence of the magnetic field, a charged particle moves on a loop enclosing a magnetic flux and thus can acquire an Aharonov–Bohm phase. In order to obtain such a closed trajectory, a ‘Mexican-hat potential’ was proposed by Kettemann, instead of the usual double-well potential [15]. The resulting flux  $\phi = \pi r^2 B$ , with  $r$  being the hat radius, proves to be smaller by several orders of magnitude than the flux  $\phi_0 = h/e$ . Even though the existence of cluster configurations of atoms or molecules containing up to  $N = 200$  units which contribute to collective tunnelling were reported (see for example [16] and references therein), it is improbable that such an effect can be extended to an amount of the order of  $N \sim 10^5$ . A detailed discussion of this aspect has been presented in [17]. The combination of the Aharonov–Bohm effect with a flip–flop configuration of the two interacting TLSs has been proposed to explain the dependence of the echo amplitudes on the applied magnetic field [18]. A different modality to consider the occurrence of an apparent flux phase having the correct order of magnitude was proposed by Würger [19]. The mechanism proposed consists of pair of coupled TLSs and a non-linear coupling to the external voltage. Still, the required degeneracy of the nearby TLS is not in accordance with the known distribution for the tunnel splitting. Another possibility for the formation of closed loops was proposed by Le Cochec [9], namely a jagged and uneven potential landscape between the two wells.

To conclude, the ‘orbital’ models can provide an explanation for some of the magnetic field effects by considering the flux dependence of the tunnelling splitting. Unfortunately, some assumptions have been made which cannot be reconciled with the standard features of the tunnelling model.

The spin models [20] provide an alternative mechanism for the observed magnetic properties, especially for the polarization echoes [21]. A direct coupling between the nuclear spin of the tunnelling entities and the applied magnetic field is considered. We can easily notice that the multi-component glasses used in the echo experiments contain one or several kinds of atoms that carry a nuclear quadrupole. For example, in the case of BaO–Al<sub>2</sub>O<sub>3</sub>–SiO<sub>2</sub> we find the abundant isotopes <sup>27</sup>Al ( $I = 5/2$ ) and less frequent <sup>137</sup>Ba ( $I = 3/2$ ); for the boro-silicate we find the abundant isotope <sup>11</sup>B ( $I = 3/2$ ). However, the echo experiments convincingly demonstrate the role of the nuclear quadrupole moments in glasses. According to this model the magnetic properties should not be measured in materials whose nuclei contain no finite quadrupole moment. Until now no counter-example has been reported. Moreover, recently the role of the quadrupole moments has been systematically studied and confirmed by the echo experiments in ordinary C<sub>3</sub>H<sub>8</sub>O<sub>3</sub> and deuterated C<sub>3</sub>D<sub>8</sub>O<sub>3</sub> glycerol [22].

The question of whether or not the nuclear quadrupole should be responsible for the magnetic field dependence of the dielectric constant seems to be natural. Relevant experiments involve temperature where the thermal energy is significantly larger than the quadrupole splitting. For thermal tunnelling systems, with TLS splitting ( $E$ ) much larger than the quadrupole splitting, it has been proven already that the resonant, or van Vleck, susceptibility is rather insensitive to the nuclear quadrupole motion [23]. For such systems the relaxation or Debye part of the susceptibility shows a large magnetic field dependence [24]. By considering TLSs with small splitting comparable to the nuclear quadrupole energies and using a numerical estimation, a pronounced dependence of the electric permittivity of the applied magnetic field was obtained [25].

In this paper we have studied the magnetic field dependence of the dielectric susceptibility and the heat capacity in cold glasses, taking into account the quadrupole effects. The general expressions in the mentioned cases are complex; however, we have obtained the thermodynamic behaviour in the small and large magnetic field limits. In section 2 we introduce the nuclear spins in the frame of the TLS. The resulting magnetic field dependent part of the susceptibility is given in section 3, using the perturbation expansion for two different regimes, namely small and large magnetic fields. We end this section with a discussion of our results and a comparison with available experimental data and previous calculations. In section 4, the specific heat has been calculated in three separate terms, namely TLS, angular momentum and nuclear spin parts. Finally, the conclusion is presented with a discussion of our results and a comparison with available experimental data [26].

## 2. TLS with a nuclear spin

The standard TLS can be described as a particle or a small group of particles moving in an effective double-well potential (DWP). At very low temperatures, only the ground states corresponding to the two wells are relevant. Using a pseudo-spin representation the Hamiltonian of such a TLS is written

$$H_{\text{TLS}} = \frac{1}{2}\Delta_0\sigma_x + \frac{1}{2}\Delta\sigma_z, \quad (1)$$

where  $\sigma_z$  is the reduced two-state coordinate,

$$\sigma_z = |L\rangle\langle L| - |R\rangle\langle R|.$$

The eigenvalues of  $\sigma_z$  are  $\pm 1$ , labelling the localized states in the two wells (left well  $L$ , right well  $R$ ), while the tunnelling matrix is taken into account by

$$\sigma_x = |L\rangle\langle R| + |R\rangle\langle L|.$$

We denote by  $\Delta$  the energy off-set at the bottom of the wells, and by  $\Delta_0$  the tunnel matrix element. According to the randomness of the glassy structure, the energy difference between the two wells

$$E = \sqrt{\Delta_0^2 + \Delta^2}, \quad (2)$$

has a broad distribution. The energy off-set and the tunnelling matrix element obey a distribution law

$$\mathcal{P}(\Delta, \Delta_0) = \frac{P_0}{\Delta_0}, \quad (3)$$

where  $P_0$  is a constant. It is useful to define new spin operators using the relations

$$\begin{aligned} \sigma_x &= u\sigma_E - w\sigma_A, \\ \sigma_z &= w\sigma_E + u\sigma_A, \end{aligned} \quad (4)$$

such that the TLS Hamiltonian becomes diagonal

$$H_{\text{TLS}}^D = \frac{E}{2}\sigma_E. \quad (5)$$

We used the notations

$$u = \frac{\Delta_0}{E}, \quad w = \frac{\Delta}{E} \quad (6)$$

which satisfy  $u^2 + w^2 = 1$ .

For the moment there is no rigorous theory for tunnelling in glasses. It is assumed that atoms or groups of atoms participate in one TLS. As we mentioned before, in the case of the multi-component glasses, one or several of the tunnelling atoms carry a nuclear magnetic dipole and an electric quadrupole. When the system moves from one well to another, the atoms change their positions by a fraction of an ångström.

We can describe the internal motion of the nuclei by a nuclear spin  $\mathbf{I}$  of absolute value  $I^2 = \hbar^2 I(I+1)$ . This is related to a magnetic dipole moment  $g\mu_N \mathbf{I}/\hbar$ , where  $g$  is the Landé factor and  $\mu_N \approx 5 \times 10^{-27} \text{ J T}^{-1}$  is the nuclear magneton. The magnetic dipole couples to an external magnetic field  $\mathbf{B} = B\mathbf{e}_B$  and gives rise to a Zeeman term

$$H_Z^{(1)} = -\epsilon_z e_B \cdot I, \quad (7)$$

where the frequency  $\epsilon_z = g\mu_N B/\hbar$  is directly proportional to  $B$ .

For a nucleus with spin quantum number  $I \geq 1$  the charge distribution  $\rho(\mathbf{r})$  is not isotropic. Besides the charge monopole, an electric quadrupole moment can be defined with respect to an axis  $\mathbf{e}$

$$Q = \int d^3r [3(\mathbf{r} \cdot \mathbf{e})^2 - r^2] \rho(\mathbf{r}). \quad (8)$$

This can couple to an electric field gradient (EFG) at the nuclear position, expressed by the curvature of the crystal field potential. The potential describing this coupling reads as [27]

$$V_Q = \frac{-eQ}{I(2I-1)} [V_{11}I_1^2 + V_{22}I_2^2 + V_{33}I_3^2]. \quad (9)$$

The bases used here ( $e_1, e_2, e_3$ ) are the principal axes of the tensor  $V_{ij}$  which describes the electric field gradient, and  $e$  is the electron charge. According to the Laplace equation the potential obeys  $V_{11} + V_{22} + V_{33} = 0$ . If we define the asymmetry parameter  $\eta = \frac{V_{22} - V_{33}}{V_{11}}$ , the quadrupole potential can be expressed as

$$V_Q = \epsilon_q [3I_1^2 + \eta(I_2^2 - I_3^2) - I^2], \quad (10)$$

where we denote by  $\epsilon_q = \frac{-eQV_{11}}{4I(2I-1)}$  the quadrupole coupling constant.

### 3. Static dielectric susceptibility

Our purpose is to determine the magnetic field dependent part of the static dielectric susceptibility for a TLS coupled with a nuclear quadrupole. Therefore, we have to consider the interaction of the dipole operator  $(1/2)P\sigma_z$  with an external electric field  $F$ . The dipole moment arises from the relative motion of partial charges related to the atoms forming the tunnel system. This interaction will be described by

$$V_F = (P \cdot F)\sigma_z. \quad (11)$$

If we denote

$$Z = \text{Tr}\{e^{-\beta H}\}, \quad (12)$$

the partition function of the system, where  $H$  is the total Hamiltonian; the trace, denoted later by  $\langle \dots \rangle$  involves two-state and spin variables. Here  $\beta = \frac{1}{k_B T}$ ,  $k_B$  is the Boltzmann constant and  $T$  is temperature. We can express the susceptibility as

$$\hat{\chi} = -\left\langle \frac{\partial^2 f}{\partial F^2} \right\rangle, \quad (13)$$

where  $f = -\frac{1}{\beta} \ln Z$  represents the free energy. The statistical average also implies an integration over the two parameters of the TLS, the energy off-set and the tunnelling matrix element, according to the distribution law given by equation (3), as well as an integration over the nuclear quadrupole parameters; this step, denoted by  $\overline{(\dots)}$ , will be discussed at the end of the calculation. We can write the partition function in terms of the energy levels  $E_{i,m}$  (eigenvalues of the total Hamiltonian)

$$Z = \sum_{i=L/R} \sum_{m=-I}^I e^{-\beta E_{i,m}}. \quad (14)$$

Therefore, the susceptibility contains three terms:

$$\begin{aligned} \chi^{(1)} &= -\frac{1}{Z} \sum_{i,m} e^{-\beta E_{i,m}} \frac{\partial^2}{\partial F^2} E_{i,m}, \\ \chi^{(2)} &= \frac{\beta}{Z} \sum_{i,m} e^{-\beta E_{i,m}} \left[ \frac{\partial}{\partial F} E_{i,m} \right]^2, \\ \chi^{(3)} &= -\frac{\beta}{Z^2} \left[ \sum_{i,m} e^{-\beta E_{i,m}} \frac{\partial}{\partial F} E_{i,m} \right]^2. \end{aligned} \quad (15)$$

The eigenenergies ( $E_{i,m}$ ) of the full Hamiltonian cannot be obtained analytically in the general case because the Zeeman and nuclear quadrupole terms do not commute. In this respect we will consider two different regimes where we can obtain the analytic results in a perturbation framework. In order to develop a perturbation expansion we will consider two separate cases, a small external magnetic field and a large one, respectively.

#### 3.1. Small magnetic field

We begin with the case in which the Zeeman term is smaller than the nuclear quadrupole potential,  $\varepsilon_z \ll \varepsilon_q$ , thus the Zeeman term is treated as a weak perturbation. If we consider the simplest symmetric case,  $\eta = 0$ , then the nuclear quadrupole potential in the  $I_1$  basis has the following representation:

$$H_q = \left[ \varepsilon_q^L \mathcal{V}_{I,m} \left( \frac{1 + \sigma_z}{2} \right) + \varepsilon_q^R \mathcal{V}_{I,m} \left( \frac{1 - \sigma_z}{2} \right) \right] |I, m\rangle \langle I, m|, \quad (16)$$

where we have defined  $\mathcal{V}_{I,m} = 3m^2 - I(I+1)$ , and  $\varepsilon_q^{L/R}$  as the quadrupole coupling constant in the left and the right well. Therefore, in the  $\sigma_E$  basis we can write

$$H_q = [\varepsilon_q \mathcal{V}_{I,m} + \varepsilon'_q \mathcal{V}_{I,m} (w\sigma_E + u\sigma_A)] |I, m\rangle \langle I, m|. \quad (17)$$

Here we have defined  $\varepsilon_q = \frac{1}{2}(\varepsilon_q^L + \varepsilon_q^R)$  and  $\varepsilon'_q = \frac{1}{2}(\varepsilon_q^L - \varepsilon_q^R)$ . We can split the total Hamiltonian into two parts,

$$H = H^{(0)} + \delta H, \quad (18)$$

where the unperturbed part is

$$H^{(0)} = \left[ \frac{E}{2} \sigma_E + \varepsilon_q \mathcal{V}_{I,m} - m\varepsilon_z \cos \theta' \right] |I, m\rangle \langle I, m|, \quad (19)$$

and the perturbation term is

$$\delta H = [(\varepsilon'_q \mathcal{V}_{I,m} + PF)(w\sigma_E + u\sigma_A)] |I, m\rangle \langle I, m| - \alpha_-^{m'} \varepsilon_z |I, m\rangle \langle I, m+1| - \alpha_+^{m'} \varepsilon_z |I, m\rangle \langle I, m-1|. \quad (20)$$

We have defined

$$\alpha_{\pm}^{m'} = \sqrt{I(I+1) - m(m \pm 1)} \sin \theta' e^{-[\pm i\phi']}, \quad (21)$$

where  $\theta'$  and  $\phi'$  are the direction of magnetic field in the basis  $(e_1, e_2, e_3)$ ; here we have assumed that  $e_1 = e_z$ .

We apply the perturbation theory to get the second order correction for the energy levels of the system,  $E_{i,m}$ . Therefore, the first order correction is

$$\delta E_{i,m}^{(1)} = w(P \cdot F + \varepsilon'_q \mathcal{V}_{I,m}) \langle i | \sigma_E | i \rangle. \quad (22)$$

For the sake of simplicity we use  $\langle i | \sigma_E | i \rangle$  instead of  $\langle i, m | \sigma_E | i, m \rangle$ , which is independent of the magnitude of  $m$ . The second order correction of the energy reads

$$\delta E_{i,m}^{(2)} = \sum_{j,n|(j,n) \neq (i,m)} \frac{|\langle i, m | \delta H | j, n \rangle|^2}{E_{i,m}^{(0)} - E_{j,n}^{(0)}} = \varepsilon_z^2 \left[ \frac{|\alpha_+^{m'}|^2}{(2m-1)\varepsilon_q - \varepsilon_z \cos \theta'} - \frac{|\alpha_-^{m'}|^2}{(2m+1)\varepsilon_q - \varepsilon_z \cos \theta'} \right] + \frac{u^2}{E} (P \cdot F + \varepsilon'_q \mathcal{V}_{I,m})^2 \langle i | \sigma_E | i \rangle, \quad (23)$$

where  $E_{i,m}^{(0)}$  is the eigenvalue of the unperturbed part of the Hamiltonian  $H^{(0)}$ . Taking into account the second order correction of energy, the susceptibility is calculated by equation (15). By using the relations

$$\frac{\partial}{\partial F} E_{i,m}|_{F=0} = wP \langle i | \sigma_E | i \rangle,$$

and

$$\frac{\partial^2}{\partial F^2} E_{i,m}|_{F=0} = \frac{2P^2 u^2}{E} \langle i | \sigma_E | i \rangle, \quad (24)$$

we can immediately express the second term of the susceptibility

$$\chi_2 = \beta P^2 w^2. \quad (25)$$

The third term is

$$\chi_3 = -P^2 w^2 \beta t^2, \quad (26)$$

where we defined  $t = \tanh(\beta E/2)$ . The first term of the susceptibility contains the second derivative of the energy levels with respect to the electric field. After some simple algebra,  $\chi_1$  can be written as

$$\chi_1 = \frac{2P^2u^2}{ZE} \sum_m [e^{-\beta E_{d,m}} - e^{-\beta E_{u,m}}], \quad (27)$$

where  $Z = \sum_m [e^{-\beta E_{d,m}} + e^{-\beta E_{u,m}}]$ . Defining  $\Lambda_m$  as

$$\Lambda_m = \varepsilon_q \mathcal{V}_{I,m} - \varepsilon_z m \cos \theta' + \varepsilon_z^2 \left( \frac{|\alpha_+^m|^2}{(2m-1)\varepsilon_q - \varepsilon_z \cos \theta'} - \frac{|\alpha_-^m|^2}{(2m+1)\varepsilon_q - \varepsilon_z \cos \theta'} \right),$$

one can write that

$$E_{i,m}|_{F=0} = \left[ \frac{E}{2} + \frac{u^2}{E} (\varepsilon_q' \mathcal{V}_{I,m})^2 \right] \langle i | \sigma_E | i \rangle + \Lambda_m. \quad (28)$$

The partition function can be written as  $Z = \sum_m \cosh[\frac{\beta E}{2} + \frac{u^2 \beta}{E} (\varepsilon_q' \mathcal{V}_{I,m})^2] e^{-\beta \Lambda_m}$ . Then the first term of the susceptibility becomes

$$\chi_1 = \frac{2P^2u^2}{ZE} \sum_m \sinh \left[ \frac{\beta E}{2} + \frac{u^2 \beta}{E} (\varepsilon_q' \mathcal{V}_{I,m})^2 \right] e^{-\beta \Lambda_m}. \quad (29)$$

If at this point we assume  $\varepsilon_q' = 0$  and  $B = 0$  we will simply recover the result of the standard TLS model,  $\chi_1 = \frac{2P^2u^2 t}{E}$ .

**3.1.1. Thermal expansion.** The experiments we are addressing are performed at temperatures between a few tens and a few hundreds of millikelvin, where the thermal energy  $k_B T \sim 10^{-24}$  J is a few orders of magnitude larger than the Zeeman term (at  $B = 1$  T for example  $\mu_N B / k_B \sim 0.4$  mK) or the quadrupole coupling (echo experiments suggested that  $\varepsilon_q \sim \varepsilon_z$  at 200 mT, so smaller than 1 mK). We can conclude that the parameters  $\beta \varepsilon_z$ ,  $\beta \varepsilon_q$  are small, which justifies an expansion of  $\chi_1$  and the exponential factors like  $e^{\pm \beta \varepsilon_{z,q}}$ . Carrying out such an expansion in the thermal energy, we can easily show that

$$\begin{aligned} \Delta \chi_1(B) &\simeq \frac{2P^2u^4}{(2I+1)E^2} (1-t^2) \varepsilon_q'^2 \beta^2 \varepsilon_z^2 \\ &\times \sum_m \left[ \frac{|\alpha_+^m|^2}{(2m-1)\varepsilon_q - \varepsilon_z \cos \theta'} - \frac{|\alpha_-^m|^2}{(2m+1)\varepsilon_q - \varepsilon_z \cos \theta'} \right]. \end{aligned} \quad (30)$$

In order to complete the calculation of the magnetic field dependent part of the static susceptibility we need to perform the integral over the TLS parameters, considering the distribution function defined in equation (3); for this regime ( $\varepsilon_z \ll \varepsilon_q$ ), which can be rewritten as

$$\Delta \chi_1(B) \simeq \frac{2P^2 \kappa \xi}{(2I+1)} \beta^3 \varepsilon_q'^2 \frac{\varepsilon_z^2}{\varepsilon_q}, \quad (31)$$

where  $\kappa = [\frac{1}{2I+1} (\sum_m \mathcal{V}_{I,m}^2) (\sum_m \alpha_m) - \sum_m \mathcal{V}_{I,m}^2 \alpha_m]$ ,  $\alpha_m = \sum_m [\frac{|\alpha_+^m|^2}{(2m-1)} - \frac{|\alpha_-^m|^2}{(2m+1)}]$ , and  $\xi$  is a (non-magnetic-field-dependent) constant which comes from averaging over the TLS parameters, which depends on the magnitude of the lower and upper bound of the two-level splitting,  $\Delta_{0\min}$  and  $E_{\max}$ . It can be easily shown that in the case of the same EFG in the two wells ( $\varepsilon_q^L = \varepsilon_q^R$ ) the dependence of the static dielectric susceptibility on the magnetic field will vanish.



### 3.2. Large magnetic field

Let us denote by  $(e_1, e_2, e_3)$  the basis in the left well and by  $(e'_1, e'_2, e'_3)$  the corresponding one in the right well. If we suppose that  $e'_3 \parallel e_3 \parallel \hat{z}$  and  $e'_1, e_1$  make an angle  $\theta$  with each other [25], we can write the total Hamiltonian in the basis of the operator  $I_3 = I_z$  in the following form:

$$\begin{pmatrix} H^L + (\frac{\Delta}{2} + PF) \cdot \mathbf{1} & \frac{\Delta_0}{2} \cdot \mathbf{1} \\ \frac{\Delta_0}{2} \cdot \mathbf{1} & H^R - (\frac{\Delta}{2} + PF) \cdot \mathbf{1} \end{pmatrix} \quad (32)$$

where  $\mathbf{1}$  is the unit matrix of rank  $(2I + 1)$ .  $H^L = V_Q^L + H_Z^{(1)L}$  and  $H^R = V_Q^R + H_Z^{(1)R}$  are defined for particles in the left and right wells, respectively. The state in each well ( $L$  or  $R$ ) can be characterized by  $|\psi_{I,m}^{L(R)}\rangle = |L(R)\rangle \otimes |I, m\rangle$  and consequently the matrix element of the Hamiltonian is defined by the following equation:

$$H_{m,n}^{L(R)} = \langle \psi_{I,m}^{L(R)} | H^{L(R)} | \psi_{I,n}^{L(R)} \rangle. \quad (33)$$

After some simple algebra (see for example [28]) we can easily show that for  $H_{m,n}^R$  we obtain

$$\begin{aligned} H_{m,n}^R &= -\epsilon_z \cos \theta'' m \delta_{m,n} + \epsilon_q^R \delta_{m,n} \Upsilon_{m,m} - \epsilon_z \sin \theta'' [e^{i\phi''} \alpha_-^m \delta_{m+1,n} + e^{-i\phi''} \alpha_+^m \delta_{m-1,n}] \\ &+ \frac{\epsilon_q^R}{4} \delta_{m-2,n} \Upsilon_{m,m-2} e^{2i\theta} + \frac{\epsilon_q^R}{4} \delta_{m+2,n} \Upsilon_{m,m+2} e^{-2i\theta}, \end{aligned} \quad (34)$$

where  $\theta''$  and  $\phi''$  define the direction of magnetic field in the basis  $(e_1, e_2, e_3)$ ;  $\alpha_-^m$  and  $\alpha_+^m$  denote

$$\alpha_{\pm}^m = \sqrt{I(I+1) - m(m \pm 1)}, \quad (35)$$

and

$$\begin{aligned} \Upsilon_{m,m} &= \left[ \frac{1}{2} I(I+1) - \frac{3}{2} m^2 + \frac{\eta}{2} [I(I+1) - 3m^2] \right], \\ \Upsilon_{m,m-2} &= \frac{1}{4} [(3-\eta)[I(I+1) - (m-1)(m-2)]^{\frac{1}{2}} [I(I+1) - m(m-1)]^{\frac{1}{2}}], \\ \Upsilon_{m,m+2} &= \frac{1}{4} [(3-\eta)[I(I+1) - (m+1)(m+2)]^{\frac{1}{2}} [I(I+1) - m(m+1)]^{\frac{1}{2}}]. \end{aligned} \quad (36)$$

In a similar manner we can obtain the matrix element of the Hamiltonian for the particles in the left well  $H_{m,n}^L$  by taking  $\theta = 0$  and changing  $\epsilon_q^R \mapsto \epsilon_q^L$  in equation (34).

We will use the perturbation approach assuming that  $\epsilon_z \gg \epsilon_q$  (here for satisfying the perturbation condition we consider the special magnetic direction in a way that  $\theta'' \ll \frac{\pi}{2}$ ). It proves to be useful to change the representation of the Hamiltonian into the basis of the spin operators  $\sigma_E$  and  $\sigma_A$ . In this basis we split the total Hamiltonian of system as  $H_0^D + \delta V$ , where  $H_0^D$ , the unperturbed part, will now contain the diagonal matrix elements of the total Hamiltonian except those coming from  $V_F$ ; therefore, the perturbation,  $\delta V$ , will contain the non-diagonal terms of the Hamiltonian together with  $V_F$ . We find that

$$H_0^D = \sum_{m=-I}^I \left[ \frac{E}{2} \sigma_E - (m\epsilon_z \cos \theta'') \mathbf{1} \right] |I, m\rangle \langle I, m|,$$

and

$$\langle m | \delta V | n \rangle = \epsilon_{mn} + \eta_{mn} \omega \sigma_E + \eta_{mn} u \sigma_A. \quad (37)$$

We denote  $\epsilon_{mn}^{L/R} = \langle m | \delta V^{L/R} | n \rangle$  and

$$\epsilon_{mn} = \frac{1}{2} (\epsilon_{mn}^L + \epsilon_{mn}^R), \quad \eta_{mn} = \frac{1}{2} (\epsilon_{mn}^L - \epsilon_{mn}^R). \quad (38)$$

The diagonal part of the perturbation term is

$$V_F^D = (\omega \sigma_E + u \sigma_A) P \cdot F. \quad (39)$$

The first order correction of the energy level will be obtained as

$$\delta E_{i,m}^{(1)} = wP \cdot F \langle i | \sigma_E | i \rangle + \varepsilon_{mm} + \eta_{mm} w \langle i | \sigma_E | i \rangle, \quad (40)$$

and the second order one will be

$$\begin{aligned} \delta E_{i,m}^{(2)} = & P^2 F^2 u^2 \sum_{j \neq i} \frac{1}{E_{i,m}^{(0)} - E_{j,m}^{(0)}} + P F u^2 \sum_{j \neq i} \left[ \frac{|\eta_{mm} \langle j | \sigma_A | i \rangle|^2}{E_{i,m}^{(0)} - E_{j,m}^{(0)}} + \text{C.C.} \right] \\ & + \sum_{j \neq i} \frac{|\eta_{mm}|^2 u^2}{E_{i,m}^{(0)} - E_{j,m}^{(0)}} + \sum_{n \neq m} \frac{|\langle i | \sigma_E | i \rangle \eta_{mn} w + \varepsilon_{mn} \mathbf{1}|^2}{E_{i,m}^{(0)} - E_{i,n}^{(0)}}. \end{aligned} \quad (41)$$

Since we have assumed that  $\delta V$  does not contain any diagonal term,  $\varepsilon_{mm} = \eta_{mm} = 0$ , the corrections to the energy levels simplify to

$$\delta E_{i,m}^{(1)} = wP \cdot F \langle i | \sigma_E | i \rangle, \quad (42)$$

and

$$\delta E_{i,m}^{(2)} = \frac{P^2 F^2 u^2}{E} \langle i | \sigma_E | i \rangle + \sum_{n \neq m} \frac{|\langle i | \sigma_E | i \rangle \eta_{mn} w + \varepsilon_{mn} \mathbf{1}|^2}{E_{i,m}^{(0)} - E_{i,n}^{(0)}}. \quad (43)$$

For the large magnetic field regime we calculate the first and the second derivatives of  $E_{i,m}$  with respect to the electric field

$$\frac{\partial}{\partial F} E_{i,m} |_{F=0} = wP \langle i | \sigma_E | i \rangle,$$

and

$$\frac{\partial^2}{\partial F^2} E_{i,m} |_{F=0} = \frac{2P^2 u^2}{E} \langle i | \sigma_E | i \rangle. \quad (44)$$

We can easily express the susceptibility

$$\chi^{(1)} = \frac{2P^2 u^2}{ZE} \sum_m [e^{-\beta E_{d,m}} - e^{-\beta E_{u,m}}]. \quad (45)$$

Here  $Z = \sum_m [e^{-\beta E_{d,m}} + e^{-\beta E_{u,m}}]$  is the partition function, and  $E_{d,m}$  and  $E_{u,m}$  are defined by

$$E_{i,m} |_{F=0} = \frac{E}{2} \langle i | \sigma_E | i \rangle - m \cos \theta'' \varepsilon_z + \sum_{n \neq m} \frac{|\langle i | \sigma_E | i \rangle \eta_{mn} w + \varepsilon_{mn} \mathbf{1}|^2}{E_{i,m}^{(0)} - E_{i,n}^{(0)}}. \quad (46)$$

**3.2.1. Thermal expansion.** Let us remember here that the thermal energy exceeds by a few orders of magnitude the Zeeman energy and the nuclear quadrupole potential. For this reason we can carry out a thermal expansion in terms of  $\beta \varepsilon_q$ ,  $\beta \varepsilon_z$  and find

$$e^{-\beta E_{i,m}} = e^{-\beta \left( \frac{E}{2} \langle i | \sigma_E | i \rangle \right)} \left[ 1 + m \cos \theta'' \beta \varepsilon_z - \beta \sum_{n \neq m} \frac{|\langle i | \sigma_E | i \rangle \eta_{mn} w + \varepsilon_{mn} \mathbf{1}|^2}{-\cos \theta'' \varepsilon_z (m - n)} \right] + O(\beta^2 \varepsilon^2). \quad (47)$$

After some algebra, we can easily write

$$\chi^{(1)} = \frac{2P^2 u^2}{E} t + \frac{2\beta P^2 u^2}{E(2I + 1)} (1 - t^2) \sum_{m,n \neq m} w \frac{\eta_{mn}^* \varepsilon_{mn} + \eta_{mn} \varepsilon_{mn}^*}{\cos \theta'' \varepsilon_z (m - n)} + O(\beta^2 \varepsilon^2). \quad (48)$$

Similar to the small magnetic field regime, from equations (15) and (44) we can straightforwardly show that

$$\chi^{(2)} + \chi^{(3)} = P^2 w^2 \beta (1 - t^2). \quad (49)$$

The magnetic field dependent correction of the susceptibility will be

$$\delta\chi(B) = \frac{4\beta P^2}{E(2I+1)}(1-t^2)wu^2 \sum_{m,n \neq m} \frac{\text{Re}[\eta_{mn}^* \epsilon_{mn}]}{\cos \theta'' \epsilon_z(m-n)} + O(\beta^2 \epsilon^2). \quad (50)$$

If we set  $\epsilon_q = 0$  and assume that  $I = 0$ , we recover once more the result of the standard TLS model,

$$\chi_{\text{TLS}} = \frac{2P^2 u^2}{E} t + P^2 w^2 \beta (1-t^2). \quad (51)$$

By averaging over the TLS parameters according to the distribution function (3), we obtain

$$\delta\chi(B) = \frac{4\beta P^2 \xi'}{(2I+1)} \sum_{m,n \neq m} \frac{\text{Re}[\eta_{mn}^* \epsilon_{mn}]}{\cos \theta'' \epsilon_z(m-n)} + O(\beta^2 \epsilon^2), \quad (52)$$

where  $\xi'$  is the numeric (non-magnetic-field-dependent) constant, which depends on the magnitude of  $E_{\text{max}}$  and  $\Delta_{0\text{min}}$ . We can easily see that  $\delta\chi(B) \sim \frac{\beta \bar{\epsilon}_q^2}{\epsilon_z}$ , where  $\bar{\epsilon}_q^2 \propto [(\epsilon_q^L)^2 - (\epsilon_q^R)^2]$ .

As we expected from the beginning, if we neglect the phase difference between the nuclear moment in the wells and if we assume that the EFGs in both wells are the same ( $\epsilon_q^L = \epsilon_q^R$ ), then we do not find any magnetic dependence in the second order perturbation (equations (31) and (52)).

### 3.3. Discussion on the field dependence of dielectric susceptibility

In this section we have addressed the question of whether or not the coupling of the two-state coordinate and the nuclear spin variables through the nuclear quadrupole potential in an inhomogeneous crystal field can be taken into account as the source of the magnetic field dependence of the dielectric susceptibility.

Analysing the existent data regarding the dielectric susceptibility as a function of the magnetic field of different oxide glasses and mixed crystals [6–10], we can notice along with a pronounced bump around 200 mT some irregular oscillations. The curvature of the susceptibility changes its sign a few times up to 5 T magnetic field. The amplitude of the real part of the susceptibility seems to be about 10% of the TLS susceptibility, corresponding to approximately  $10^{-4}$  of the dielectric constant. Relevant experiments involve temperatures from tens of millikelvin to a few hundred. We can observe that in this range the dielectric constant varies with the inverse temperature,  $1/T$ .

The present work reconsiders the static dielectric susceptibility of a glassy systems in an external magnetic field, implementing a perturbation approach for the energy levels. Starting from the coupling of the nuclear quadrupole with the tunnelling system, calculations have been performed for two different regimes, i.e. small and large external magnetic fields with respect to the nuclear quadrupole potential. We have found that the magnetic field dependent part of the susceptibility in both regimes is the following:

$$\Delta\chi_{\text{small}}(B) \sim \beta^2 \epsilon_z^2 \frac{\epsilon_q^2}{\epsilon_q},$$

$$\Delta\chi_{\text{large}}(B) \sim \beta \frac{\bar{\epsilon}_q^2}{\epsilon_z}.$$

As it is obvious from the above results, the dielectric susceptibility depends on the magnetic field in the second order correction of the perturbation scheme via the Zeeman and the nuclear

quadrupole terms. This correction is directly the result of different electric field gradients (EFGs) in the two wells. In the small magnetic field regime the correction increases with  $B^2$  while it decreases inversely with field ( $1/B$ ) in the large magnetic field. The dependence on the temperature was put into evidence in agreement with the existent experimental data. The magnetic field dependence will disappear in the second order corrections if we take the same EFG in both wells. We would like to recall that the previous existing perturbation calculations [23] which considered the same EFG provides a magnetic field dependence of the dielectric susceptibility only to the fourth order of expansion.

The theoretical approach we have used does not allow us to calculate the static dielectric susceptibility for the case where the applied magnetic field has such values such that the Zeeman energy and the nuclear quadrupole have similar magnitudes,  $\epsilon_z \simeq \epsilon_q$ . But for this regime we know [23] that the magnetic field dependent part of the static susceptibility  $\chi$  is proportional to the small ratio  $[(\epsilon_z \epsilon_q)/T^3]^2$ , this ratio being of the order of  $10^{-6}$  (one millionth part) for  $T \sim 1$  K.

We have to mention here that our calculation provides only the static limit ( $\omega = 0$ ) of this contribution.

The dynamical susceptibility of a simple TLS was expressed by Jäckle [29],

$$\chi_{\text{TLS}}^{\text{rel}}(\omega) = \left\langle P^2 w^2 \frac{\beta}{\cosh^2(\beta E/2)} \frac{i\gamma}{\omega + i\gamma} \right\rangle, \quad (53)$$

where  $\gamma$  is the relaxation rate. Now, by considering the coupling of the nuclear quadrupole to the tunnelling motion, the dynamic susceptibility proves to be more complicated, but it can be generally expressed as [24]

$$\chi^{\text{rel}}(\omega) = \left\langle P^2 w^2 \frac{\beta}{\cosh^2(\beta E/2)} \sum_m A_m \frac{i r_m}{\omega + i r_m} \right\rangle, \quad (54)$$

where  $r_m$  represents the eigenvalues of the relaxation rate matrix, and  $A_m$  the corresponding amplitudes ( $m = 0, \dots, 2I+1$ ). In the limit of  $\omega \rightarrow 0$ , an approximation of the susceptibility is provided by equations (31) and (52) for the two considered regimes. Recalling that the relevant experiments are performed at frequencies in the range of kilohertz, and that the relaxation rates should be extremely small quantities (details will be give elsewhere [24]), we can conclude that such a limiting case cannot provide a proper explanation for the existent experimental data.

The method we developed cannot provide a plausible value for the relative magnitude of the magnetic field dependent part with respect to the simple TLS susceptibility. We do not find oscillations of the dielectric susceptibility at magnetic fields of the order of 1 T.

#### 4. The heat capacity

Our purpose is to find the magnetic field dependence of the heat capacity in multi-component glasses. To the best of our knowledge there are not many experimental data for heat capacity of (nonmagnetic) glasses in the presence of magnetic field. Therefore, we assume that the tunnelling particles have an angular momentum,  $J$  (intrinsic angular momentum such as electron spin). Then, we must add an additional term to the Hamiltonian, which in the presence of magnetic field leads to

$$H_Z^{(2)} = -\epsilon'_z J_z. \quad (55)$$

The energy scale is  $\epsilon'_z = g_E \mu_B B / \hbar$ ,  $\mu_B$  is the Bohr magneton and  $g_E$  is the electronic Landé factor. Here we have neglected the spin-spin interaction in the Hamiltonian. We have also

assumed that the quantization axis of angular momentum,  $J_z$ , is in the direction of  $e_B$  (for more detail, see the appendix). The total Hamiltonian of the system is then

$$H = H_{\text{TLS}} + H_Z + H_Q.$$

Here  $H_Z = H_Z^{(1)} + H_Z^{(2)}$  (defined in equations (7), (55)) and

$$H_Q = \left(\frac{1 + \sigma_z}{2}\right)V_Q^L + \left(\frac{1 - \sigma_z}{2}\right)V_Q^R, \quad (56)$$

where  $V_Q^{R(L)}$  is defined in equation (10) for the particles in the right (left) well [23]. The heat capacity is expressed by the following relation:

$$C_v = \frac{1}{k_B T^2} \left\langle \frac{\partial^2 \ln Z}{\partial \beta^2} \right\rangle, \quad (57)$$

where  $Z$  is the partition function, which is a sum over the TLS, spin and angular momentum degrees of freedom. If we neglect the phase difference between the nuclear moments in the two wells (taking  $\theta = 0$ ) and assume that the EFGs in both wells are the same ( $\epsilon_q^R = \epsilon_q^L = \epsilon_q$ ), then the partition function can be written in the following form:

$$Z = Z_{\text{TLS}} S S', \quad (58)$$

where

$$\begin{aligned} Z_{\text{TLS}} &= \text{Tr}\{e^{-\beta H_{\text{TLS}}}\} = 2 \cosh(\beta E/2), \\ S &= \text{Tr}\{e^{-\beta(H_z^{(1)} + H_Q)}\}, \\ S' &= \text{Tr}\{e^{-\beta H_z^{(2)}}\}. \end{aligned} \quad (59)$$

Therefore, according to equations (57) and (58), we find that

$$C_v = \frac{1}{k_B T^2} \left\langle \frac{\partial^2 \ln Z_{\text{TLS}}}{\partial \beta^2} + \frac{\partial^2 \ln S}{\partial \beta^2} + \frac{\partial^2 \ln S'}{\partial \beta^2} \right\rangle = C_{\text{TLS}} + C^{(I)} + C^{(II)}. \quad (60)$$

By some calculations and after averaging over TLS parameters, we can easily show that (see for example [1])

$$C_{\text{TLS}} = \frac{\pi^2}{6} P_0 k_B^2 T \propto T, \quad (61)$$

which is independent of the magnetic field. The Zeeman contribution of the tunnelling particle's angular momentum ( $J$ ) in the partition function is simplified to the following form:

$$S' = \sum_{m'=-J}^J e^{\beta m' \epsilon'_z} = \frac{\sinh[\beta \epsilon'_z (J + \frac{1}{2})]}{\sinh[\frac{\beta \epsilon'_z}{2}]}. \quad (62)$$

Thus the Zeeman contribution of the angular momentum in the specific heat is given by

$$\begin{aligned} C^{(II)} &= \frac{P'}{k_B T^2} \left\langle \frac{\partial^2 \ln S'}{\partial \beta^2} \right\rangle = \frac{k_B \beta^2 \epsilon_z'^2}{4} \\ &\times P' \left[ \text{csch}^2\left(\frac{\beta \epsilon'_z}{2}\right) - (2J + 1)^2 \text{csch}^2\left(\frac{\beta \epsilon'_z (2J + 1)}{2}\right) \right], \end{aligned} \quad (63)$$

where  $P'$  is the concentration of intrinsic angular momentum times a constant which comes from the averaging over the TLS parameters.

Finally, the contribution of the remaining term of the heat capacity to the specific heat,  $C^{(I)}$ , is given by

$$C^{(I)} = k_B \beta^2 \left\langle \frac{\partial^2 \ln S}{\partial \beta^2} \right\rangle, \quad (64)$$

where  $S = \text{Tr}\{e^{-\beta H'}\}$  and  $H' = H_z^{(1)} + H_Q$ .

As we have mentioned in the previous section the thermal energy exceeds by a few orders of magnitude the Zeeman energy and the nuclear quadrupole potential. Therefore,  $\beta\epsilon_z$  and  $\beta\epsilon_q$  are very small quantities, consequently  $\beta H' \ll 1$ . Therefore, equation (64) is approximated by

$$\begin{aligned} C^{(I)} &= k_B \beta^2 \frac{\partial^2}{\partial \beta^2} \ln \left[ \text{Tr} \left( 1 - \beta H' + \frac{\beta^2}{2} H'^2 + \dots \right) \right] \\ &= k_B \frac{\beta^2}{2I+1} \left[ \text{Tr}(H'^2) - \frac{\text{Tr}(H')^2}{2I+1} \right] + O(\beta^3 \epsilon^3), \end{aligned} \quad (65)$$

and finally we arrive at the following expression:

$$C^{(I)} = P'' k_B \beta^2 [\gamma_1 \epsilon_z^2 + \gamma_2 \epsilon_q^2] + O(\beta^3 \epsilon^3), \quad (66)$$

where  $P''$  is the concentration of the nuclear spin times a constant which comes from the averaging over the TLS parameters. Here  $\gamma_1$  and  $\gamma_2$  are numerical constants which have been defined by (see the appendix)

$$\begin{aligned} \gamma_1 &= \frac{1}{2I+1} \sum_m [m^2 \cos^2 \theta' + \sin^2 \theta' [I(I+1) - m^2]], \\ \gamma_2 &= \frac{1}{2I+1} \sum_m \left[ \Upsilon_{m,m+2}^2 + \Upsilon_{m,m-2}^2 - \frac{\Upsilon_{m,m}^2}{2I+1} \right]. \end{aligned} \quad (67)$$

Equation (66) shows a quadratic dependence on the magnetic field. In the large field regime  $C^{(II)}$  contributes as a constant value to the whole specific heat. Thus, the field dependence of the specific heat in the high magnetic fields comes from  $C^{(I)}$ , which shows a quadratic behaviour versus the magnetic field. However, for the small field regime we have  $\epsilon_z \ll \epsilon'_z$ , which makes  $C^{(II)}$  have the dominant effect ( $C^{(I)} \ll C^{(II)}$ ). It can be shown that when the magnetic field is small the leading term of  $C^{(II)}$  is given by the following expression:

$$C^{(II)} \simeq P' k_B \beta^2 \epsilon_z^2 \frac{J(J+1)}{3}, \quad (68)$$

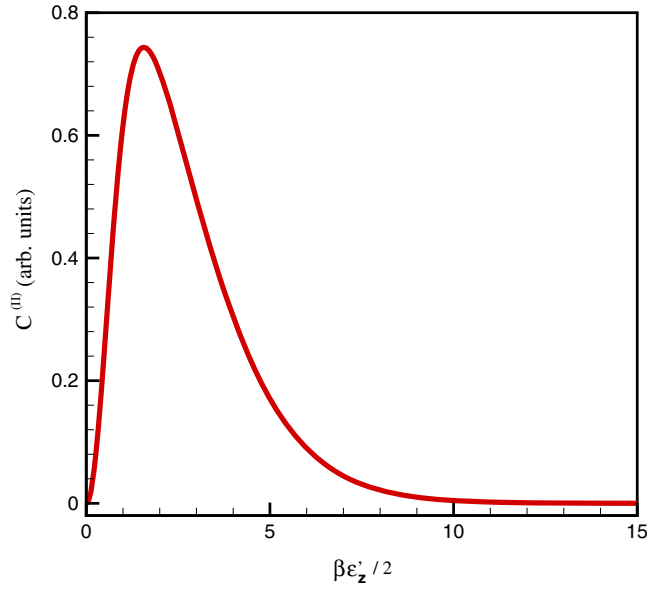
which shows  $B^2$  behaviour for low magnetic fields.

#### 4.1. Discussion of the field dependence of the heat capacity

In this section, we have studied the magnetic field dependence of the heat capacity of glasses at low temperature. We have assumed that the tunnelling particles carry both an angular momentum and a nuclear spin. So, the magnetic field couples to both terms.

We have examined our results numerically for arbitrary angle of EFG and different phases between the quadrupole moments on each well. The final results are not influenced if we only consider the simple case of  $\theta = 0$  and considering that  $\epsilon_q^R = \epsilon_q^L = \epsilon_q$ . Thus, we have reported our results in the mentioned special case where we can find an analytical expression for the specific heat behaviour. In this respect, the total partition function of the system can be split into the multiplication of three separate terms. Therefore, the heat capacity is composed of three terms, the TLS contribution ( $C_{\text{TLS}}$ ), the nuclear ( $C^{(I)}$ ) and the angular momentum ( $C^{(II)}$ ) parts. The magnetic field dependence will arise from  $C^{(I)}$  and  $C^{(II)}$ .

We have introduced the generalized Hamiltonian, which takes into account the mentioned degrees of freedom. The direction of external magnetic ( $\theta', \phi'$ ) field has been considered arbitrary with respect to the EFG direction of each TLS. However, the outcome will be averaged over this spherical angle, which can be contracted as a constant to the final results. We have calculated the contribution of  $C_{\text{TLS}}$  and  $C^{(II)}$  exactly while the effect of nuclear spin ( $C^{(I)}$ ) has been treated in a thermal expansion up to the second order corrections.



**Figure 1.** Magnetic field (thermal) variation of the  $C^{(II)}$ .  
(This figure is in colour only in the electronic version)

The different scale of Zeeman energy for the nuclear moment and the angular momentum of the tunnelling entity is roughly  $\epsilon_z/\epsilon'_z \approx 10^{-3}$ . Thus, for low magnetic fields  $\beta\epsilon'_z < 10$  the dominant effect comes from the angular momentum part ( $C^{(II)}$ ). Our calculation shows that at low magnetic field this contribution is proportional to the square of magnetic field,  $C^{(II)} \simeq B^2$  (equation (67)). However,  $C^{(II)}$  reaches a maximum at  $\beta\epsilon'_z \simeq 3$  and then decreases to zero for  $\beta\epsilon'_z > 20$  (see figure 1).

For low magnetic fields ( $B < 10$  T) our result predicts a Schottky-like peak in the specific heat of glasses which have nonzero angular momentum for the tunnelling entity, or at least the sample has some impurity with nonzero  $J$  (angular momentum). This result is in agreement with the recent experimental observations for Duran, AlBaSi [31] and Suprasil [26]. The data show explicitly an upward increase of the specific heat versus the magnetic field for  $B < 0.5$  T and a monotonic decrease for  $B > 1$  T at  $T < 1$  K.

We have also predicted that for high magnetic fields ( $\beta\epsilon'_z > 20$ ) the dominant contribution to the specific heat comes from the nuclear moments, which shows a quadratic dependence on the field (equation (66)).

In this paper we have studied the special case where the impurities have only the intrinsic angular momentum part. For the general case of impurities (for example electrons) with angular momentum,  $J > \frac{1}{2}$  (spin + orbit), we should take into account the effect of spin-orbit interaction, which means that at zero magnetic field the levels  $m' = -J, \dots, J-1, J$  may have different energies due to the crystal field splitting.

However, if we consider  $J = 0$ , which means we have a pure glass without magnetic impurities like BK7, the nuclear quadrupole term is the only dominant term in the specific heat. So, the present calculations predict a quadratic dependence on the magnetic field, and  $1/T^2$  dependence on temperature. As far as we know there is no experimental result for this case. Therefore it might be a good suggestion for future experiments.

## 5. Summary

Based on the polarization echo experiments [22] we believe that the nuclear quadrupole model should provide the rightful explanation for the intriguing behaviour of the dielectric properties of glasses. A relaxation spectrum rather sensitive to the orientation of the nuclear quadrupoles is providing a relaxation susceptibility strongly dependent on the applied magnetic field [24], driving us to the conclusion that the observed magnetic properties of glasses might have a relaxation origin. Other effects like non-linearities with the voltage [30], or cooperative TLS behaviour might as well be found to contribute.

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## Appendix. Generalized TLS Hamiltonian with both nuclear spin and angular momentum

The aim of this appendix is to find the generalized TLS Hamiltonian with both nuclear spin and angular momentum. In a similar way as leading to equation (32), we can write the total Hamiltonian as

$$\begin{pmatrix} H^L + \frac{\Delta_0}{2} \cdot \mathbf{1} & \frac{\Delta_0}{2} \cdot \mathbf{1} \\ \frac{\Delta_0}{2} \cdot \mathbf{1} & H^R - \frac{\Delta_0}{2} \cdot \mathbf{1} \end{pmatrix}, \quad (\text{A.1})$$

where  $\mathbf{1}$  is the unit matrix of rank  $(2I + 1) \times (2J + 1)$ .  $H^L = V_Q^L + H_Z^L$  and  $H^R = V_Q^R + H_Z^R$  are defined for particles in the left and right wells, respectively. The state in each well ( $L$  or  $R$ ) can be characterized by  $|\psi_{[I,m],[J,m']}\rangle = |L(R)\rangle \otimes |I, m\rangle \otimes |J, m'\rangle$  and consequently the matrix element of the Hamiltonian is defined by the following equation:

$$H_{mm',nn'}^{L(R)} = \langle \psi_{[I,m],[J,m']}\rangle^{L(R)} | H^{L(R)} | \psi_{[I,n],[J,n']}\rangle^{L(R)}. \quad (\text{A.2})$$

After some simple algebra we can easily show that for  $H_{mm',nn'}^R$  we obtain

$$\begin{aligned} H_{mm',nn'}^R = & \{ \epsilon_q^R [\Upsilon_{m,m-2} e^{2i\theta} \delta_{m-2,n} + \Upsilon_{m,m+2} e^{-2i\theta} \delta_{m+2,n}] \\ & - \epsilon_z \sin \theta' [e^{i\phi'} \alpha_-^m \delta_{m+1,n} + e^{-i\phi'} \alpha_+^m \delta_{m-1,n}] \\ & + [-m' \epsilon_z' - \epsilon_z \cos \theta' m + \epsilon_q^R \Upsilon_{m,m}] \delta_{m,n} \} \delta_{m',n'}, \end{aligned} \quad (\text{A.3})$$

where  $\theta'$  and  $\phi'$  define the direction of magnetic field in the basis  $(e_1, e_2, e_3)$ ;  $\alpha_{\pm}^m$  and  $\Upsilon_{m,m'}$  are given by equations (35) and (36). In a similar manner we can obtain the matrix element of the Hamiltonian for the particles in the left well  $H_{m,n}^L$  by taking  $\theta = 0$  and changing  $\epsilon_q^R \mapsto \epsilon_q^L$  in the above equation.

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