EFFECTS OF A WEAK MAGNETIC FIELD ON THE THERMAL AND ELECTRICAL PROPERTIES OF DIELECTRIC GLASSES AT LOW TEMPERATURES

Giancarlo Jug

Dipartimento di Fisica e Matematica, Università dell'Insubria
Via Valleggio 11, 22100 Como, Italy,
and INFN-Sezione di Pavia, Italy
giancarlo.jug@uninsubria.it

The physics of dielectric glasses at low temperature is interesting, in that some universal behaviour appears to be independent of the composition of the materials. For example, the heat capacity $C_p(T)$ displays the characteristic behaviour, below T~1 K:

$$C_p(T)=\gamma_{2LS}T^{1.2}+\gamma_{phon}T^3, \quad (1)$$

where the (quasi-)linear behaviour is ascribed to the presence of two-level tunnelling systems (2LS) weakly distributed in the amorphous silicate network and the cubic term is due to virtual-crystal long-wavelength phonons as in the perfect crystal [1].
More importantly, the physics of low-temperature silicate glasses is useful, in that temperature sensors can be made out of thick-films fabricated by the sol-gel technique [2] and exploiting the characteristic and universal logarithmic temperature dependence of the dielectric constant

$$\varepsilon(T)= \varepsilon(T_0)\pm A_\pm \log (T/T_0), \quad (2)$$

where $T_0(\omega)$ is a characteristic frequency-dependent temperature [3]. Experimentally, the electric capacitance thus decreases logarithmically for $T< T_0$ and increases again for $T> T_0$. The above behaviour is explained only qualitatively by the 2LS approach, in which charged tunnelling particles are assumed to move in a one-dimensional (1D) asymmetric double-well potential and where at low temperatures, effectively, only the two lowest-lying states are assumed to contribute.

The sub-Kelvin operating temperatures make the above thermometric property rather useful for studies of low-temperature physics in the presence of external magnetic fields (e.g. quantum Hall Effect physics or nanostructure physics), except that in some glasses a very unusual and remarkably enhanced (or depressed) dependence in both $C_p(T)$ both $\varepsilon(T)$ and has been observed recently [4]. The response of a structural dielectric glass to weak magnetic fields (where the enhancement is largest, with a maximum for B~0.01 T) is a very puzzling and potentially useful phenomenon, because the glass is devoid of itinerant charges or (almost) devoid of magnetic impurities. A theory for the explanation of this puzzling phenomenon is presented here, with some preliminary results for $\varepsilon(T,B)$ and a full theory for C_p (T,B).

Some earlier theoretical explanations for the effect of the magnetic field have been presented, but lead to no comprehensive explanation. The earliest proposal was for a novel quantum-coherent state of interacting 3D tunnelling systems coupled to the magnetic field via the Aharonov-Bohm (AB) effect [5]; however, this theory was shown to be flawed by the fact that the coherence is destroyed by disorder [6]. More recently, the theory effort is focusing on the coupling of the magnetic field to the tunnelling particles' double-well potential via the intermediary of the nuclear dipolar magnetic moments coupled in turn to the nuclear electric quadrupole moments (sensitive to the electric field gradients) [7]. This theoretical approach explains qualitatively the magnetic-field's polarization echo effects in these glasses [8], but implies no B-effect on $\varepsilon(T,B)$ at all [9]. Neither of the main theoretical proposals addresses the B-effect on C_p (T,B), which are also significant [10,11] and cannot be ascribed solely to the trace paramagnetic impurities (Fe).

Our theory takes the first steps from the unusual C_p (T,B) data for the multi-component silicate glasses a-AlO_3-BaO-SiO_2 and the a-borosilicate 'Duran' [10], data that complete the early study on the a-borosilicate Pyrex [11]. It appears that at B=0 already Eq. (1) is severely violated for these glasses and that for small B (~0.1 T) a maximum deviation of about 10% from the B=0 curve is attained, the law (1) to be recovered gradually as the field is increased to B~3 to 8 T (depending on the material). Paramagnetic impurities cannot explain this behaviour, so one must envisage a new type of 3D tunnelling systems in these glasses, coexisting with the standard 2LS. This comes from the fact that multi-component glasses have both network-forming (NF) and network-modifying (NM) species (NM atoms hiding around in pockets of the amorphous NF matrix) [12] and thus a simple 2LS description (albeit generalised to 3D [5]) does no longer apply to these materials. The standard 2LS tunnelling model makes use of a reduced pseudo-spin-1/2 Hamiltonian,

$$H_{2LS}=-\tfrac{1}{2}\Delta_0\sigma_x - \tfrac{1}{2}\Delta\sigma_z, \tag{3}$$

with $\sigma_{x,z}$ Pauli matrices, where Δ is roughly the two well's energy asymmetry and Δ_0 the barrier transparency (or tunnelling parameter), and of a probability distribution of these parameters of the form (independent of Δ)

$$P(\Delta_0, \Delta)=P_0/\Delta_0, \tag{4}$$

with P_0 a material-dependent constant. This can still be used to model the NF tunnelling atoms, whilst for the NM tunnelling species present in the multi-component glasses we can adopt, e.g., a reduced pseudo-spin-1 Hamiltonian, sensitive now to the magnetic field too via AB-coupling and defined by the matrix: [13]

$$H_{3LS}= \begin{matrix} E_1 & D_0e^{i\varphi/3} & D_0e^{-i\varphi/3} \\ D_0e^{-i\varphi/3} & E_2 & D_0e^{i\varphi/3} \\ D_0e^{i\varphi/3} & D_0e^{-i\varphi/3} & E_3 \end{matrix} \tag{5}$$

Here $D_0 \sim a\hbar\Omega\, e^{-b\hbar\Omega/V_0}$ (for a soft tunnelling potential; a, b are constants, Ω a single-well frequency and V_0 the barrier height) is large and positive, $\varphi=2\pi\Phi(\mathbf{B})/\Phi_0$ is the AB-phase associated to a flux $\Phi(\mathbf{B})$ threading the triangular region joining the three potential wells' minima. This Hamiltonian takes the NM atomic species' 'soft dynamics' into account, when $V_0 \approx \hbar\Omega$. One more, and very important ingredient of the present theory is a modification of the probability distribution for the parameters D_0 (3D barrier transparency) and E_1, E_2, E_3 (the wells' anisotropies):

$$P(D_0, E_1, E_2, E_3)=P^*\delta(E_1+E_2+E_3)/[D_0(E_1^2+E_2^2+E_3^2)], \tag{6}$$

with P^* a material constant for the NM-species. The above distribution, favouring the small anisotropies tunnelling systems, gets justified on the basis of the fact that nano-crystals are ubiquitous in glassy thick films prepared with the sol-gel technique [2]. These nano-crystals act as nucleation centres for the NM-species' aggregation. With the further material constants: NF-cutoffs Δ_{0min}, Δ_{0max}; NM-cutoffs D_{0min}, D_{0max}, D_{min} (where $D=(E_1^2+E_2^2+E_3^2)^{1/2}$), the concentrations x_{2LS}, x_{3LS} of the NF- and NM-species, and with the average product $S_\blacktriangle Q$ between the flux-threaded surface $S_\blacktriangle$ and the charge Q of the tunnelling system's , a systematic fit of the experimental data can be attempted, always assuming the tunnelling entities to be independent.

The results, published in [13] for the fit of the C_p (T,B) data of [10] for a-AlO_3-BaO-SiO_2 and 'Duran' and of [11] for 'Pyrex', give a very good agreement between theory and experiments in the 100 mK < T < 3 K range, when trace Fe paramagnetic impurities are also accounted for. The material parameters extracted are consistent with the assumption of the above phenomenological theory. The same parameters have been used to fit the ε(T,B) data of [4] for a-AlO_3-BaO-SiO_2 and the agreement between theory and experiments will be presented in this talk. Since the amplitude of the electric field plays also a role, the material parameters turn out to be slightly voltage-dependent. However, the peak in the dielectric constant enhancement is correctly described by this theory around B≈0.02 T, whilst the enhancement turns into a depression of $\Delta\varepsilon=\varepsilon(T,B)-\varepsilon(T,0)$ for B>0.05 T, Δε becoming actually negative . This last effect has also been observed in unpublished work by the Heidelberg group [14] and observed and published in another multi-component silicate glass by the Saclay group [15].

In summary, the present phenomenological theory explains both the heat-capacity and dielectric-constant anomalous and non-monotonic response to weak magnetic fields. The origin of these effects is the presence of both NF- and NM-species in the glasses' structures, as well as that of nano-crystals trapped in the glassy matrix and nucleated by the NM-atoms.
Useful low-temperature devices (e.g. for cryogenic particle-physics bolometry) could be made from these glassy materials, both in terms of low temperature and of weak field changes detection.

The Author thanks INFN (progetto DERBY) and MIUR (Cofin-2003) for financial support.

References
[1] W.A. Phillips, J. Low Temp. Phys. **7**, 351 (1972); P.W.Anderson, B.I. Halperin and C.M. Varma, Phil. Mag. **25**, 1 (1975)
[2] P. Strehlow, J. Non-Cryst. Solids **121**, 463 (1990); *ibid.,* Physica B **165-166**, 25 (1990); *ibid.,* Cryogenics (ICEC Suppl.) **34**, 421 (1994); see also: G. Schuster, G. Hechtfischer, D. Buck and W. Hoffmann, Rep. Prog. Phys. **57**, 187 (1994)
[3] P. Esquinazi (ed.) *Tunneling Systems in Amorphous and Crystalline Solids* (Springer, Berlin, 1998}; C. Enss, Physica, **316B-317B**, 12 (2002); *ibid.,* Adv. Sol. State Phys. **42**, 335 (2002)
[4] P. Strehlow, C. Enss and S. Hunklinger, Phys. Rev. Lett. **80**, 5361 (1998); P. Strehlow, M. Wohlfahrt, A.G.M. Jansen, R. Haueisen, G. Weiss, C. Enss and S. Hunklinger, Phys. Rev. Lett. **84**, 1938 (2000)
[5] S. Kettemann, P. Fulde and P. Strehlow, Phys. Rev. Lett. **83**, 4325 (1999)
[6] A. Langari, Phys. Rev. B **65**, 104201 (2002)
[7] A. Würger, A. Fleischmann and C. Enss, Phys. Rev. Lett. **89**, 237601 (2002)
[8] S. Ludwig, P. Nagel, S. Hunklinger and C. Enss, J. Low Temp. Phys. **131**, 89 (2003)
[9] D. Bodea and A. Würger, J. Low Temp. Phys. **136**, 39 (2004)
[10] L. Siebert, Ph.D. Thesis (Heidelberg, 2001, www.ub.uni-heidelberg.de/archiv/1601); M.Meißner and P. Strehlow, Hahn-Meitner Institute Report (Berlin, 2000, www.hmi.de/bensc/sample-env/home.html)
[11] R.B. Stephens, Phys. Rev. B **13**, 852 (1979)
[12] P. Jund, W. Kob and R. Jullien, Phys. Rev. B **64**, 1343031 (2001); J. Horbach, W. Kob and K. Binder, Phys. Rev. Lett. **88**, 125502 (2002); E. Sunyer, P. Jund, W. Kob and R. Jullien, J. Non-Cryst. Sol. **307**, 939 (2002); S. Ispas, M. Benoit, P. Jund and R. Jullien, J. Non-Cryst. Sol. **307-310**, 946 (2002); ibid., Phys. Rev. B **64**, 214206 (2002); S. Ispas, N. Zotov, S. De Wispeleare and W. Kob, cond-mat/0306332
[13] G.Jug, Phil. Mag. **84**, 3599 (2004)
[14] M. Wohlfahrt, Ph.D. Thesis (Heidelberg, 2001, www.ub.uni-heidelberg.de/archiv/1587)
[15] J. Le Cochec, F. Ladieu and P. Pari, Phys. Rev. B **66**, 064203 (2002)