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LOW TEMPERATURE PROPERTIES OF METALLIC GLASSES.

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Abstract. — The low temperature properties ($T \lesssim 1$ K) of metallic glasses are reviewed. They include specific heat, thermal conductivity, acoustic propagation mostly in superconducting glasses. They are well explained by the existence of low-energy excitations in form of tunneling two-level systems.

I. — INTRODUCTION

At low temperatures (i.e., below a few K) the behaviour of insulating non-magnetic crystals is governed by the existence of long wavelength phonons. Consequently the specific heat varies as T^3 (Debye law) and the thermal conductivity obeys also a T^3 law in the Casimir regime where the phonon mean free path is limited by the sample dimensions. The acoustic propagation is characterized by a small absorption (due to defects, impurities,...) independent of the temperature and a velocity decreasing as T^4 as a consequence of anharmonic phonon processes. In crystalline non-magnetic metals, the most important excitations are the conduction electrons. Superposed on the phonon contributions there are electronic contributions : linear in temperature for the specific heat and the thermal conductivity.

More than ten years ago it was found that the specific heat C and thermal conductivity κ of insulating glasses were different, as compared to their crystalline counterparts : an excess of C varying quasi-linearly with T and a small κ varying as T^2. The acoustic propagation was also found to be unusual. The absorption is saturable and the velocity first increases as the logarithm of the temperature. All these properties (and some others) are well described in the framework of a phenomenological model : the so-called two-level systems model (TLS model) which assumes the existence of supplementary low energy excitations strongly coupled to the phonons [1].

Six years ago it was shown that the same kind of supplementary excitations also exist in metallic glasses. The most convincing experimental evidence was provided by the acoustic experiments (logarithmic increase of the velocity and saturation of the absorption). The thermal measurements (specific

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heat and thermal conductivity) were more difficult to interpret as a consequence of the large electronic contributions which hide those from the TLS. But experiments in superconducting glasses have confirmed the existence of the TLS in metallic glasses [2].

It is worth mentioning too that other experimental techniques such as Brillouin scattering, and dielectric constant measurements, which have provided much information on insulating glasses are not possible in metallic glasses.

This paper is concerned with the low temperature properties of metallic glasses which are linked to the existence of the TLS. After a short presentation of the TLS model the experimental results which have provided evidence of the existence of the TLS in metallic glasses will be reviewed. Then the influence of the TLS on the electronic properties will be discussed and finally a microscopic description of the TLS will be given on the basis of a computer model.

II. – THE TLS MODEL.

II.1. The double-well potential.

This model was first introduced in order to explain the low temperature properties of insulating glasses [3]. Then it was extended to metallic glasses.

The basic assumption is the existence of asymmetric double-well potentials with rather low energy barriers. A particle or group of particles can move in these double-wells at low temperatures by quantum mechanical tunneling. The nature of the particle and the origin of the double-well are still unknown. Some microscopic models have been proposed to describe them. They will be discussed below. The two wells are assumed to be identical and harmonic. The two lowest energy states of the tunneling particle are separated by $E = (\Delta^2 + \Delta_0^2)^{1/2}$ where Δ is the asymmetry energy. $\Delta_0 \simeq \hbar\,\Omega\, e^{-\sigma}$ is the tunneling energy with $\Omega/2\pi$ the oscillation frequency of the particle in each well and σ is a function of the mass of the particle, the barrier height and the distance between the two wells. Because a glass is a disordered material Δ and σ have not unique values but have a distribution of values. The simplest and usual assumption is to choose flat and uncorrelated distributions for Δ and σ : $P(\Delta,\sigma) = P(\Delta)\,P(\sigma) = A$ where A is a normalization constant. For calculations it appears that E and $r = \Delta_0^2/E^2$ are more convenient variables. In that case the distribution function can be written :

$$P(E,r) = \frac{A}{2}\left(1 - r\right)^{-1/2} r^{-1} \qquad \begin{array}{c} r_m \leqslant r \leqslant 1 \\ E_{min} \leqslant E \leqslant E_{max} \end{array}$$

and 0 elsewhere.

$r_m = \Delta_m^2/E^2$, $E_{min} = \Delta_m$, $E_{max} = (\Delta_M^2 + \Delta_1^2)^{1/2}$ where Δ_m and Δ_M are respectively the minimum and maximum values of Δ_0 and Δ_1 is the maximum value of Δ .

It is now possible to obtain the density of states of the TLS.

$$n(E) = \mathcal{N} \int_{r_m}^{1} P(E,r)\, dr \simeq \frac{\mathcal{N}A}{2} \operatorname{Log} \frac{E}{\Delta_m}$$

where $\mathcal{N}$ is the total number of TLS per unit volume. A good approximation is to take $n(E) = n_0$, a constant, over a broad range of energy.

At this point it is useful to remark that the TLS can be considered to be analogous to a system of spins with $S = \frac{1}{2}$. As a consequence the formalism of the magnetism will be used in the following.

II.2. TLS-phonon and TLS-electron coupling.

An elastic strain ε modifies the parameter Δ. The strain modulation of Δ_0 is neglected. In addition Δ also depends on the local occupation of valence electron states and the conduction electrons are sensitive to changes in the electronic potential. These phenomena lead to TLS - phonon and TLS - electron coupling respectively. The hamiltonian of the TLS in a metallic glass can be written :

$$\mathcal{H} = ES_z + \left(2\gamma r^{1/2} S_x + 2\gamma(1-r)^{1/2} S_z\right)\varepsilon + \frac{1}{N}\sum_{k,q} \left(K_e r^{1/2} S_x + K_e(1-r)^{1/2} S_z\right) C_k^+ C_{k+q} \quad (1)$$

$\gamma = \partial\Delta/\partial\varepsilon$ and K_e characterize the TLS - phonon and TLS - electron couplings respectively. C_α^+ and C_α are the creation and annihilation operators respectively of an electron in the state α. N is the total number of atoms. K_e can be evaluated in a model where the electronic states are assumed to be plane waves [4]. In the following K_e and γ will be taken as parameters to be determined by experiments.

The interaction of phonons (or electrons) occurs by two mechanisms :

i. Resonant : the off diagonal terms in (1) describe the resonant transition between the two states of the TLS by absorption or emission of a phonon of energy $\hbar\omega = E$ or by scattering of an electron.

ii. Relaxational : the diagonal terms modulate the TLS splittings E . The perturbed TLS population relaxes towards equilibrium with a characteristic relaxation time T_1 by interaction with thermal phonons, conduction electrons or quasiparticles in the superconducting states.

These two interaction mechanisms lead to changes in the elastic constant resulting in contributions to the acoustic attenuation and to the acoustic velocity [5]. There are also changes in the electron scattering rates and in the corresponding resistivity.

III. – EXPERIMENTAL EVIDENCE FOR THE EXISTENCE OF THE TLS.

III.1. Specific heat.

Knowing the TLS density of states, the TLS contribution to the specific heat can be calculated. It is given by $C = \frac{\pi^2}{6} k_B^2 T n_0$ [3]. At low temperatures ($T \lesssim 1$ K) this term is preponderant in insulators. In metals it is masked by the electronic contribution varying also as T . But in superconductors this electronic contribution vanishes for $T \ll T_c$. Experiments have been performed in glassy superconductors : the first one in $Pd_{30}Zr_{70}$ where $T_c \simeq 2.6$ K [6]. At the lowest temperatures ($T \lesssim 0.3$ K) there is clearly an excess specific heat over the electronic and Debye contributions. Measurements in various superconducting glasses including Cu_xZr_{1-x} [7], $M_{24}Zr_{76}$ [8] (M = Ni, Pt, Ag, Cu), $Zr_{75}Rh_{25}$[9], have confirmed the existence of an extra contribution to the specific heat. Some typical curves are shown in figure 1. In all cases, even in the most careful experiments the excess specific heat cannot be fitted with a linear law but with a T^α law (α varying from 0.5 to 0.9).

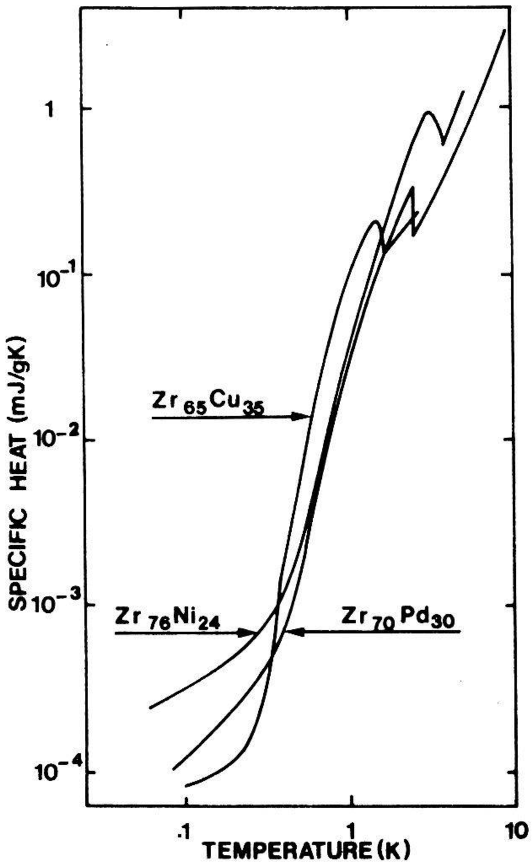

FIGURE 1. – Low temperature specific heat of three superconducting glasses : $Zr_{70}Pd_{30}$ (from [6]), $Cu_{35}Zr_{65}$ (from [7] and $Ni_{24}Zr_{76}$ (from [8]).

In conclusion, the specific heat results in glassy superconductors show the existence of supplementary low energy excitations but are not entirely consistent with a constant density of states. However it can be determined that the density of states is of the same order of magnitude as in insulating glasses (about 10^{33} erg^{-1} cm^{-3} which corresponds to 10^{17} TLS per cm^3 uniformly distributed in energy from 0 to 1 K).

III.2. Thermal conductivity.

If the thermal phonons are scattered by resonant interaction with the TLS, their mean free paths are strongly reduced. Therefore the phonon thermal conductivity is small and varies as T^2 as a consequence of the constant TLS density of states [3]. In a metallic glass the electronic thermal conductivity (varying as T) can be estimated using the Wiedemann-Franz law. A "glassy" $T^{\sim 2}$ contribution has been reported in a-PdSi [10]. More convincingly a $T^{\sim 1.9}$ thermal conductivity has been measured in various superconducting glasses, first in a $Pd_{30}Zr_{70}$ alloy [6] and then in Zr_xBe_{1-x} [11], Cu_xZr_{1-x} [7], $Ni_{24}Zr_{76}$ [12] alloys (see figure 2).

From these experiments evidence is clearly found for the existence of a broad band of TLS with nearly constant density of states and strongly coupled to the phonons. The product of the TLS constant density of states by the square of an average coupling constant γ can be estimated. The order of magnitude ($\sim 5 \times 10^7$ erg cm^{-3}) is similar to the value found in fused silica. Moreover, it has been found that the phonon thermal conductivity has a plateau [10]. This behaviour, whose origin is still not satisfactorily understood, appears quite common to all glassy materials whether metallic or insulating.

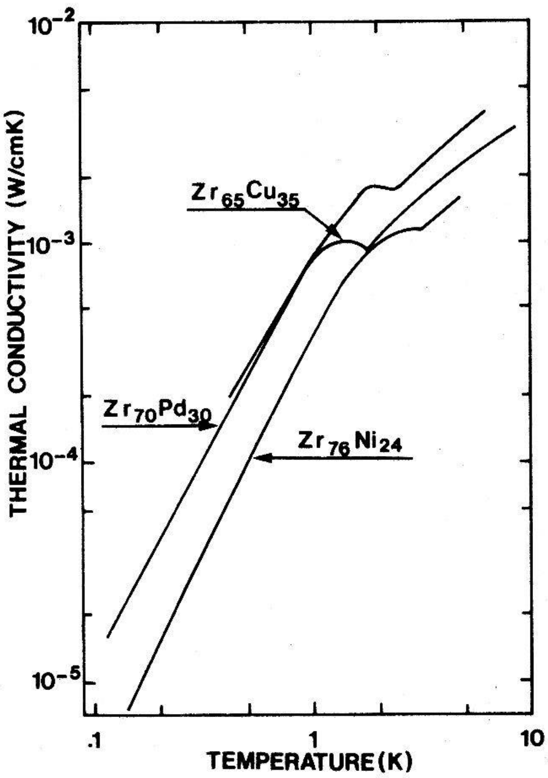

FIGURE 2. - Low temperature thermal conductivity of three superconducting glasses : $Zr_{70}Pd_{30}$ (from [6]), $Zr_{65}Cu_{35}$ (from [7]) and $Ni_{24}Zr_{76}$ (from [12]).

The specific heat and thermal conductivity measurements in superconducting glasses give also some other information concerning the electronic density of states, Debye temperature, electron - phonon coupling and superconductivity [7,13]. However, a discussion of these points is beyond the scope of this review.

III.3. Acoustic propagation.

First it is important to note that phonon - phonon interactions and electron - phonon interactions provide negligeably small contributions to acoustic absorption and velocity changes in the temperature and frequency ranges of interest here [14].

III.3.1. Resonant contribution.

The resonant interaction between TLS and an acoustic wave of frequency $\omega/2\pi$ leads to a temperature dependent change of the acoustic velocity v when $\hbar\omega \ll kT$ of the form [5] :

$$\frac{\Delta v}{v} = C \; \ell n (T/T_0) \tag{2}$$

where T_0 is an arbitrary reference temperature, $C = \mathcal{N} A \gamma^2 / 2 \rho v^2$ with ρ the density of the material. The observation of this logarithmic temperature dependence in glassy NiP [15] , PdSiCu [16] and CoP [17] was actually the first experimental evidence for the existence of TLS in metallic glasses. The related acoustic absorption is given in terms of phonon mean free path [5] :

$$\ell^{-1} = \left(1 + \Phi/\Phi_c\right)^{-1/2} \frac{\pi\omega}{v} C \; tanh(\hbar\omega/2kT) \tag{3}$$

where Φ is the acoustic flux in the sample and Φ_c a critical flux given by $\Phi_c = \hbar^2 \rho v^3 / 2 \gamma^2 T_1 T_2$ where T_2 is the transverse relaxation time of the TLS.

The first factor in equation (3) expresses the saturation of the acoustic absorption when the transition rate is so high that the relaxation of the population cannot restore the thermal equilibrium. This saturation effect is quite characteristic of the TLS model (as compared to a model of harmonic oscillators) and consequently is often considered as one of its best tests. It has been observed in glassy PdSiCu [18,19] and NiP [20]. The main difference with the insulating glasses is the order of magnitude of Φ_c $\left(\Phi_c \text{ (metals)} \simeq 10^4 \; \Phi_c \text{ (insulators)}\right)$.

This effect can be understood if the TLS - electron coupling is taken into account. This interaction provides a very efficient channel to the relaxation of the TLS at the lowest temperatures resulting in very short relaxation times and therefore a high value for Φ_c (the relaxation times are in the nanosecond range at 1 GHz and 0.1 K).

The frequency and temperature dependences predicted by equation (3) have also been observed on a PdSiCu sample [21].

III.3.2. Relaxational contribution.

This part of the attenuation can be observed when the resonant contribution is saturated at high acoustic flux. In terms of the complex changes $\delta c(\omega,T)$ of the elastic constant c_0 this contribution is given by :

$$\frac{\delta c}{c_0} = - \frac{1}{4c_0} \int_0^\infty dE \int_{r_{min}}^1 dr \; P(E,r) \; sech^2\left(\frac{\beta E}{2}\right) \frac{\beta \gamma^2 (1-r)}{1 + i\omega T_1} \tag{4}$$

The attenuation is given by $\ell^{-1} = \frac{\omega}{v} \, Im \{ \delta c / c_0 \}$ and the velocity change by $\frac{\Delta v}{v} = \frac{1}{2} \, Re \{ \delta c / c_0 \}$. In metal T_1 is given by $T_1^{-1} = (T_1)_{el}^{-1} + (T_1)_{ph}^{-1}$ where the indices refer to processes where relaxation occurs by diffusion of one conduction electron and by absorption or emission of one thermal phonon respectively. In insulating glasses where the relaxation of the TLS occurs via the direct phonon process only $\left(T_1^{-1} \propto r \gamma^2 E^3 \; coth(\beta E/2) \right)$ the relaxational acoustic attenuation at the lowest temperatures is frequency independent and increases as T^3 [22]. If the most efficient relaxation process is due to electrons $\left(T_1^{-1} \propto r K_e^2 E \coth(\beta E/2)\right)$ [19] the attenuation is predicted to be frequency independent and linear in temperature. This behaviour has been observed in NiP [20] and PdSiCu [21]. The corresponding velocity changes are small and masked by the resonant contribution.

At higher temperatures the relaxational contribution reaches a plateau i.e. an attenuation independent of the temperature and varying linearly with the frequency [22]. Moreover this law is independent of the relaxation process. This is a direct consequence of the distribution of the coupling constants. In NiP and PdSiCu this plateau is not observed [23]. It is probably masked by the low temperature side of an Arrhenius peak.

In the usual experimental conditions the relaxational contribution is not saturable. But recently saturation of the relaxational absorption has been proposed [24] in order to explain strong nonlinear effects different from the usual saturation of the resonant contribution observed in the acoustic absorption in PdSiCu and other glassy alloys [25]. These effects could originate from equalization of the TLS occupation numbers by the acoustic wave over a spectral width of the order of $kT/\hbar$.

III.3.3. Acoustic propagation in glassy superconductors.

A conclusive test of the importance of the TLS - electron coupling is to perform ultrasonic measurements in glassy superconductors because of the condensation of electrons into pairs below T_c. The TLS model has been extended to such a case and spectacular features have been predicted for the acoustic propagation near T_c partly as a consequence of a supplementary relaxation process due to excitation of quasiparticles across the superconducting energy gap by interaction with the TLS [26].

Experiments have been done on $Cu_{60}Zr_{40}$ alloys ($T_c \simeq 0.4$ K) [27,28]. The results show a strong decrease of the absorption just below T_c and a plateau above T_c (see figure 3). The sound velocity first increases as the logarithm of the temperature, passes through a maximum below T_c and through a minimum just at T_c and above T_c increases logarithmically with a slope which is one half of the slope of the $\ln T$ at low temperatures (figure 4). These variations are in qualitative agreement and partly in quantitative agreement with the predicted behaviours [26]. It is important to note that experiments in a $Pd_{30}Zr_{70}$ alloy (which were in fact the first acoustic experiments in a glassy superconductor) did not show the "anomalies" near T_c [29]. This was explained [30,31] in the framework of the TLS model as a consequence of the rather high T_c (2.6 K) and the very strong TLS electron coupling which leads to very short T_1 relaxation times of the TLS. The application of a magnetic field has also clearly shown the role of the electrons [29].

From this section, it can be concluded that acoustic experiments in metallic glasses have proved the existence of a constant density of states of TLS strongly coupled both to phonons and electrons. The differences in the behaviour as compared to the insulating glasses are consequences of the TLS - electron coupling.

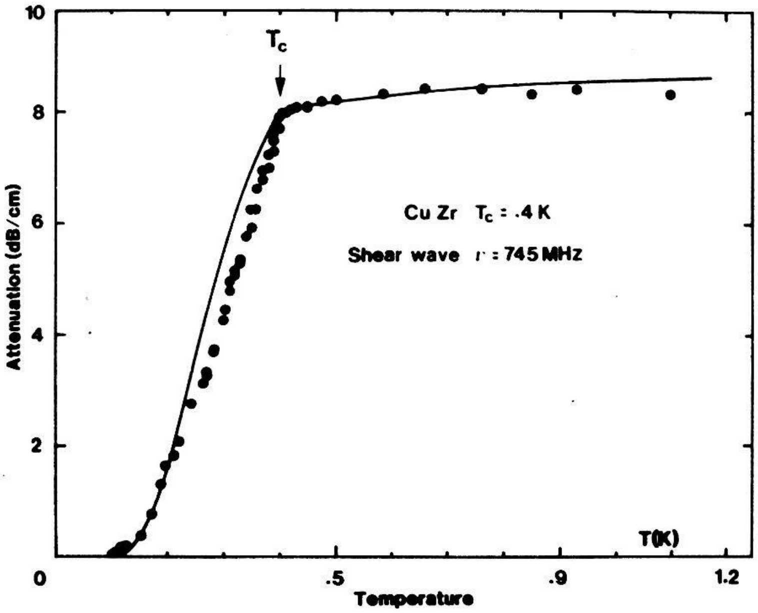

FIGURE 3. - Attenuation of transverse acoustic waves in a $Cu_{40}Zr_{60}$ superconducting glass. Solid line is a calculated curve (from [27]).

IV. - ELECTRICAL RESISTIVITY.

Because of the strong TLS - electron coupling the question arises concerning the influence of the TLS on the electronic properties. This point is not yet clear although much experimental and theoretical work has been done on the subject. From the experimental point of view the existence of a minimum and a logarithmic temperature increase of the resistivity below the minimum is now well established in some systems where the Kondo (magnetic) origin of this behaviour has been ruled out [32]. It has been suggested that this effect was due to electronic scattering by the TLS [33]. This problem has been discussed recently in detail [34]. It is clear from calcuting the lowest order inelastic

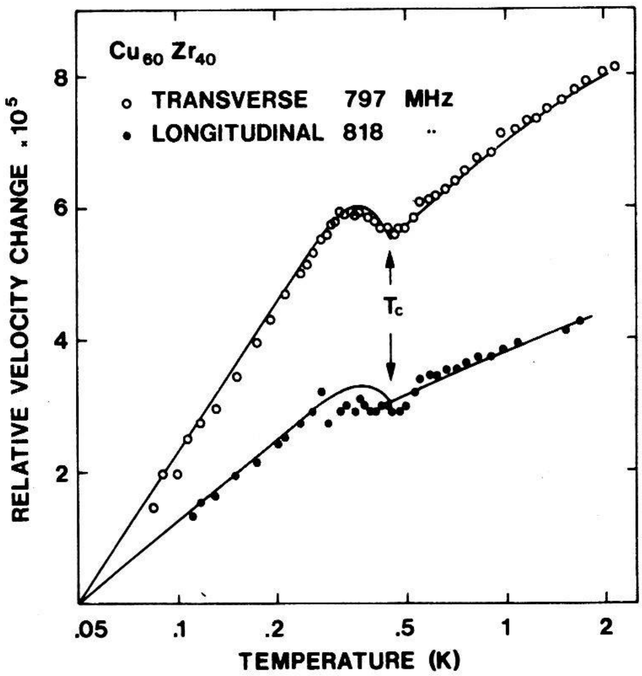

FIGURE 4. - Change in the phase velocity of longitudinal and transverse acoustic wave in a $Cu_{60}Zr_{40}$ superconducting glass. Solid lines are calculated curves (from [28]).

scattering process that the contribution to the resistivity is several orders of magnitude too low to be consistent with the experiments [4]. The possibility of many body enhancements has been discussed [34]. It has been predicted that in the case of very strong TLS - electron coupling a resistance minimum occurs.

Another interesting possibility in this field is that conduction electrons may have an attractive interaction mediated by the TLS leading to a new mechanism for superconductivity. The calculated critical temperature T_C [35] depends critically on the strength of the TLS - electron coupling and it is difficult to compare it to the actual value of T_C.

More experimental work is necessary before it will be possible to conclude that a TLS contribution to the electronic properties exists.

V. - MICROSCOPIC ORIGIN OF THE TLS.

Until now the TLS model as presented above is quite phenomenological. The question of its microscopic origin is of central importance for a complete understanding of the low temperature properties of glasses. Some microscopic descriptions of the TLS have been proposed. They concern generally a particular class of glass : silica [5] , chalcogenides [36], metals [37] and in that sense the universality of the existence of the TLS is lost. The description of the TLS in the framework of the free-volume theory [38] does not suffer this loss of generality. But the theoretical description of glass properties from this concept of free volume is not firmly established.

An interesting approach to this problem is the use of computer models to get some insight into the microscopic realization of a TLS. Such a model has been recently studied [39] for a binary metallic alloy. The possibility that a single atom can tunnel between two metastable minimum energy positions has been found. Moreover it was found that all these TLS are associated with voids in the structure and that the TLS density of states decreases during a structural relaxation process. This last finding is important. It can be related to changes in the low temperature properties of metallic glasses before and after annealing. There is experimental evidence for a reduction of the TLS density of states from thermal conductivity measurements in PdSiCu [10], ZrNi [12] and ZrCu [40] alloys. A decrease of the low temperature excess specific heat in a ZrNi [12] alloy after annealing has been also measured.

VI. - CONCLUSION.

The low temperature properties (specific heat, thermal conductivity, acoustic propagation) of metallic glasses are governed by the existence of

TLS strongly coupled to phonons and electrons. It is worth emphasizing that these properties are *-i.* quite different from those of the crystals, *-ii.* quite similar (except of course the TLS - electron coupling and its consequences) to those of insulating glasses and probably all amorphous systems.

Finally we would mention some specific points which are either not yet understood or still conjectural or disputed. The existence of a T^3 term in the specific heat larger than the usual T^3 Debye contribution which is common to all insulating glasses has not been firmly established in metallic glasses due to the lack of sound velocity measurements. This point is not understood as well as the existence of a plateau in the thermal conductivity and the rapid quasilinear decrease of the sound velocity after the maximum at a few K. These two latter features are common to all glasses whether they are metallic or insulating.

The influence of the TLS on the electronic properties (resistivity or also the possibility of a new mechanism for the superconductiving) is still an open question. But the major challenge remaining is of course to find what the microscopic origin of the TLS is.

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