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DEVIOUS STRUCTURAL RELAXATION FROM THE PURE DEBYE CHARACTER IN Fe\textsubscript{40} Ni\textsubscript{40} B\textsubscript{20} METALLIC GLASS

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Abstract - Structural relaxation by topological and chemical short-range order (TSRO and CSRO) in Fe\textsubscript{40} Ni\textsubscript{40} B\textsubscript{20} metallic glass was investigated using Young's modulus, internal friction and magnetic energy loss measurements. The vibration reed technique in the low-frequency range and the hysteresis loop method were adopted. From the frequency shifts of the absorption bands observed at different temperatures below crystallization, activation energies of 0.6 and 1.7 eV were calculated for TSRO and CSRO, respectively. Theoretical analysis revealed that short-range order taking place in metallic glasses was characterized by a single relaxation time, but with a distribution of activation energies showing itself as an asymmetric tail in the relaxation spectrum at lower temperatures. A model based on the quantization of free volume in the amorphous metallic structure was presented and was used to interpret the values and the distribution of activation energies for both types of short-range order.

Introduction -
It is presently well known /1-4/ that structural relaxations do exist in metallic glasses before the onset of crystallization. The relaxation involving local atomic shifts that do not discriminate between different types of constituent atoms gives rise to topological short-range order (TSRO). Chemical relaxation occurring at relatively higher temperatures involves atomic bonding of similar magnetic atoms, and is usually termed chemical short-range order (CSRO). Mechanical properties such as internal friction, Q\textsuperscript{-1}, and Young's modulus, E, were found to be more sensitive to relaxation by TSRO /5/, whereas magnetic properties were usually taken to trace CSRO /6/. Due to the amorphous structure of metallic glasses and the random distribution of free volume in the matrix, it is thought that the unit relaxational element might change slightly from place to place. The aim of the present work is to test the deviation of the overall relaxation from the pure Debye relaxation, and to find out whether there is a distribution in activation energy and/or relaxation time.
Experimental

Flexural vibrations of short ribbons of Fe$_{40}$ Ni$_{40}$ B$_{20}$ (*) were electrostatically traced by a micro-vibration detector. The internal friction was determined by the resonance curve method to an accuracy of 4%. Changes in the elastic modulus of the order of 0.005 could be determined from the change in the natural frequency of the ribbon. The magnetic energy loss was determined from the hysteresis loop area recorded by the oscilloscope method.

Results

(i) Internal friction and Young's modulus

The temperature dependence of internal friction, $Q^{-1}$, and Young's modulus, $E$, indicated a relaxational behaviour at temperatures slightly above 400 K showing itself as a peak in $Q^{-1}$, associated with a drop in $E$. The temperatures at which the relaxation was fully operative was found to shift to higher values as the vibration frequency increased (Fig. 1). The relation between relaxation temperature and the corresponding vibration frequency (Fig. 2) yielded an energy of 0.6 eV activating the relaxation process in action.

(ii) Magnetic energy loss

The magnetic energy loss, tan $\delta$, of the test sample was measured at different temperatures up to the crystallization. A characteristic peak in tan $\delta$ superimposed the broad peak effected by crystallization. This proved to have a relaxation origin as evidenced by the observed shift to higher temperatures as the field frequency increased (Fig. 3). The frequency shift yielded an activation energy of 1.7 eV.

Discussion

(i) Analysis of the two relaxation peaks

In view of the random close packed structure of the amorphous test sample, structural relaxation processes seemed to occur with a spectrum in activation energies and relaxation times. To interpret the present observations, we first assume that the acting processes are thermally activated with a single relaxation time. The normalized $Q^{-1}$ can thus be expressed in the form /8/,

$$\frac{Q^{-1}}{Q^{-1}_{M}} = \text{sech} \left( \frac{-H}{k(T_{M} - T^{-1})} \right),$$

where $Q^{-1}_{M}$ is the peak value of internal friction occurring at temperature, $T_{M}$. Accordingly, a plot of $Q^{-1}_{M} / Q^{-1}$ versus $(T_{M} - T^{-1})$ would yield a symmetric curve about $T = T_{M}$ for a single relaxation process. Substituting in equation (1) by the activation energy, $H = 0.6$ eV, here deduced for TSRO, values of $Q^{-1}_{M}$ were calculated and the dashed curve in Fig. 4 resulted. It could be seen from Fig. 4 that the experimental data fitted the theoretical curve at relatively high temperatures while it showed a large low temperature asymmetry. This asymmetric tail of the relaxation spectrum was previously observed /9,10/ and was commonly considered an intrinsic property of the amorphous state. Applying the same analysis to the absorption band detected magnetically, a similar asymmetric behaviour of the relaxation band was found (Fig. 5). Although the origin of this relaxation by CSRO is different from that detected mechanically, yet the asymmetric tail observed in both relaxation spectra implied that the activation energy for short-range order fluctuations from place to place within the matrix despite the uniqueness of the relaxation time.

(ii) Relaxation by TSRO

Considering the microscopic model for atomic tilting previously presented by the authors /11/, which is based on a quantization of free volume in metallic glasses, the last conclusion could be interpreted as follows. The random distribution of the free

(*) The samples were received from Max-Planck institut für metallforschung, Stuttgart, West Germany.
(111) Relaxation by CSRO

Bonding of the similar ferromagnetic atoms (Fe-Fe, Ni-Ni) in the metallic glass Fe$_{40}$Ni$_{40}$B$_{20}$ seemed to form the origin of CSRO. Here also, the relaxation mechanism was controlled by free volume migration associated with atomic tilting and shifting. This was confirmed by the suggestion of Chen and Egami [6] that the kinetics of the CSRO are determined by the availability of the free volume in the nearby locality. The high activation energy (1.7 eV) observed for the CSRO might be inferred to necessary atomic displacements that bring together similar ferromagnetic atoms. The fluctuation of this value from place to place could be also explained in terms of the presence of free volume in different amounts varying from place to place, thus modifying the energy barrier for atomic displacements. The decrease in the chemical potential by bonding two similar magnetic atoms together forms the motive force for the migration of vacies. A unit migration of a quantum of void might not be sufficient to bring about this bonding process. Several vacy jumps might be needed to produce one required link, i.e., one magnetic dipole. On account of previously calculated energy for TSRO (0.6 eV) which is a result of a single vacy shift, one

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Fig. 1 - Temperature dependence of the modulus $E/E_U$, and the normalized internal friction peak after being removed from its background, for Fe$_{40}$Ni$_{40}$B$_{20}$ metallic glass. The plots show how the modulus and internal friction vary with temperature for different frequencies.

Fig. 2 - Natural frequency of the sample versus the inverse temperature at which the relaxation is fully operative.
Fig. 3 - Temperature dependence of magnetic energy loss, tan δ, for Fe_{40}Ni_{40}B_{20} metallic glass.

Fig. 4 - Internal friction versus the inverse temperature for the first relaxation process. The dashed curve represents a single relaxation of activation energy, \( H = 0.6 \) eV.

Fig. 5 - Magnetic energy loss versus the inverse temperature for the second relaxation process. The dashed curve represents a single relaxation of activation energy, \( H = 1.7 \) eV.

would estimate that the unit relaxation process of CSRO would require, on the average, three vacancy shifts needing about, 1.7 eV.
References -