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CONDUCTIVITY MEASUREMENTS ON UHV DEPOSITED AMORPHOUS SILICON

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Abstract.- Conductivity has been investigated on UHV deposited a-Si thick films as a function of spin concentration. Results are analysed using Mott's VRH theory and can be described by laws of the form \( \sigma = \sigma_0 \exp \left(-\frac{\Theta_0}{T}\right) \). The variations of \( \Theta_0 \) and \( \sigma_0 \) with spin concentration are presented. Relations between ESR signal linewidth, longitudinal relaxation time \( T_1 \), and conductivity are investigated. In addition to a spin-lattice relaxation rate process of the form \( T_1^{-1} \propto T \), we evidence another contribution which is proportional to dc conductivity.

Introduction

The need of a study of conductivity as a function of spin concentration (N) using pure enough samples to avoid contributions of impurity effects exists because of the dispersion of experimental results obtained on a-Si and a-Ge and of the difficulty of their description using one-phonon-assisted hopping theories (I). Possible relations between conductivity and ESR have already been pointed out (2-5). It appears that the temperature dependent linewidth broadening may originate not only from a process associated with transport but also from other contributions that must be investigated. We report here a study of conductivity as a function of spin concentration and a study of the ESR longitudinal relaxation time \( T_1 \), that we try to correlate to the temperature dependent ESR linewidth and to the conductivity.

Experimental

Thick a-Si films (0.6-1 \( \mu \)) are deposited in UHV under conditions that were previously described (5). The spin concentration is varied by annealing treatments performed in UHV in the range 100-500°C of samples deposited on unheated substrates; the spin concentration is a decreasing function of annealing temperature \( T_A \) and varies at most by a factor of 3. Aluminum contacts are deposited in another vacuum system after very short air exposure. Distance between electrodes is about 150 \( \mu \). We get perfectly ohmic contacts at any measuring temperature which is determined using a calibrated silicon diode. Errors are of the order 0.1 K in the range 77-300 K and may reach 0.3 K near 40 K.

Conductivity and spin concentration

One observes on fig. 1 a strong dependence of conductivity on \( T_A \) (i.e. N) as it has already been reported. RT absolute values of conductivity are similar to those reported by Knotek (evaporated a-Si) and Lewis (sputtered a-Si). There exist an apparent linear regime in the \( T_1^{-1/2} \) representation in the range 50-150 K and deviations toward lower conductivities at higher temperatures. At temperatures below 60 K we observe deviations toward higher conductivities that are only slightly above possible experimental errors so that we do not consider their reality as firmly established. A characteristic feature concerns the slope of lines which obviously varies with \( T_A \). To investigate the reality of the apparent agreement of the linear part with a \( T_1^{-1/2} \) regime, we use a linear regression method to determine the value of the parameter \( n \) which gives the best fit in a log \( \sigma \) representation. Fig. 2 shows that the best fits correspond for all samples to \( n \) values close to 4.
Fig. 1 - Conductivity versus $T^{-1/4}$ measured on UHV deposited a-Si films annealed in UHV at temperature $T_A$ after deposition on unheated substrates.

Fig. 2 - A linear regression method is applied to $\log_{10} \sigma = f(T/\Gamma)^{1/4}$. Normalised mean square sums are plotted versus $n$. Best fits correspond to $n$ values close to 4.

The effect of a possible temperature dependence of the preexponential term is also shown on fig. 2 where the typical shift of the position of the minimum of curves arising from $\sigma \tau = \sigma_0 \exp \left(-\frac{T_0}{T}\right)$ representation (Mott), is indicated. Shifts being of the order of the dispersion of curves it appears difficult to draw conclusions about the temperature dependence of the preexponential term.

Assuming a constant density of states at Fermi level ($N_F$) we have used Mott's expression for $\Sigma$ and calculated both $N_F$ and the localisation length $\alpha$.

Even if we adjust the phonon term of the preexponential factor to get reasonable values for $N_F$, we obtain too large (2 orders of magnitude) a variation of $N_F$ and unreasonable $\alpha$ values which do not seem to describe the data. Of course, the use of $T_0$ values alone, assuming the value of one term ($N_F$ or $\alpha$'), allows to get consistent values for the other (for instance $\alpha' = 3$ Å and $T_0 = 2 \times 10^8$ K give $N_F \approx 4 \times 10^{15}$ eV$^{-1}$ cm$^{-3}$). If we assume the existence of a simple proportionality relation between $N_F$ and $N_S$, which means that only the intensity and
not the shape of the density of states varies with $N$, we can compare the variations of $T$ and $\sigma$ with $N$ to those expected from Mott's VRH theory. The data give $\sigma \propto N^{0.5}$ which is quite different from $\sigma \propto N^{1/3}$ that is predicted by Mott's expression. The dependence of $T_0$ on $N$ is shown on fig. 3. It reveals a smaller variation than the inverse proportionality which is expected, and may be related to the fact that $T_0$ not only depends on the value of the density of states at Fermi level but also on its shape that may change with spin concentration. The conclusion of these results is that Mott's formula for VRH including the preexponential term cannot describe the data. However $T_0$ values and dependence on $N$ do not seem in contradiction with the expression of the exponential term derived in one phonon assisted VRH theories.

Longitudinal relaxation time and conductivity

The full ESR linewidth broadening with temperature was attributed to the relaxation associated with hopping (2). This assignment is supported by the fact that it is possible theoretically to evidence processes for carriers jumps which can contribute both to transport and spin lifetime. For instance a theory of spin flip induced by the jump itself has been given by Movaghar et al (4). However the origin of the ESR linewidth broadening with $T$ has not been proved and one can suggest the possible effect of other mechanisms as usual (independant on spins jumps) spin-lattice relaxation. The study of the temperature dependence of the longitudinal relaxation time $T_1$ is expected to bring new information on the mechanisms involved. In a-Si, the presence of short $T_1$'s (samples with large $N$) and the nature of the spin system can make the use of the continuous saturation method difficult. The $X$ band ESR lineshapes are always almost Lorentzian. This shows that inhomogeneous contributions (e.g. g value distribution) must have comparatively small influence. This is no more the case for $Q$ band lines (P.A. Thomas unpublished). Moreover a spread of $T_1$ values exists as previously reported (4). Although the presence of even a small inhomogeneous contribution to the line requires in principle a more complex treatment, we have analysed $X$ band data using the theory of continuous saturation for homogeneous lines, We do not expect that this approximation influences the temperature dependence of $T_1$. However one can get, because of the spread in $T_1$'s some errors on its absolute values. $T_1$ versus $T$ is shown on fig. 4. We get a large dependence of $T_1$ on $T_0$ (i.e. $N$) which is not discussed here, and a $T_1 \propto T^\alpha$ law for $T < 150$ K; deviations toward lower $T_1$ appear for $T > 150$ K. Gourdon et al (6) have obtained similar results using a different method. We can verify, assuming a single homogeneous spin packet that the observed linewidth broadening ($\Delta H$) correspond to the present $T_1$ values : fig. 5 shows good agreement, considering experimental errors, between observed and calculated linewidths (the same $1,25 T$ correcting factor was used to fit the 3 curves). We think as Gourdon et al., that the $T_1 \propto T^\alpha$ dependence can be attributed to a one-phonon process, but in contrast to these authors, we do not consider that it necessarily implies the presence of large spin clusters. At least two possibilities can be envisaged to describe the deviations above 150 K: a) a two-phonon process which is expected to give $T_1 \propto T^{2\alpha}$ and to manifold above $T \sim 0.3 N^1$ that does not disagree with experiment; b) a contribution to the relaxation rate
Fig. 5 - The lifetime broadening calculated with $T_1$ values (points) fits the temperature dependent part of the peak to peak linewidth (lines).

$T_1$ related to spin jumps. To test these possibilities we assume two independent relaxation rates: $T_1^{-1} = T_{1L}^{-1} + T_{1h}^{-1}$, where $T_{1h} \propto T$. We can then calculate $T_{1h}$ from data in the range 150-300 K. The conductivity is plotted versus $T_{1h}$ on fig. 6 which evidences a high correlation and a nearly one to one relation over 3 orders of magnitude. We think that this result establishes, beyond the origin of the deviation to $T_1^{-1} \propto T$ above 150 K, that spins are mobile and participate to transport. The data show that at least a large part of the spin population can relax via this process, whereas only spins with energy near $E_F$ contribute to transport; this does not imply that all spin states have an energy close to $E_F$, as the whole spin population is exchange-coupled (the mean exchange value is larger than the linewidth at X band; P.A. Thomas unpublished). The consequences of this $T_{1h} \propto \sigma$ law will be presented elsewhere.

Conclusion.
Our data show that spin centers are directly involved in transport. This is supported by the evidence of a contribution to spin lattice relaxation rate which is proportional to dc conductivity and by the strong dependence of transport parameters on spin concentration. The analysis of conductivity which is well described on the range 60-150 K by $\ln \sigma \propto \left( T / T_0 \right)^{1/4}$ where $T_0$ is spin concentration dependent, encounters serious difficulties if one uses the preexponential term given by Mott's VRH theory.

References

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