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NUCLEAR QUADRUPOLEAR RELAXATION IN LIQUID Ga-Te ALLOYS

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Abstract. The nuclear spin relaxation rate was measured in liquid $\text{Ga}_c\text{Te}_{1-c}$ alloys on As as an impurity atom. A TDPAD technique was used on the $6 \mu\text{s}$ $^{73\text{m}}\text{As}$ isomer. The range $1 \geq c \geq 0.2$ was studied, with $T=1110 \text{ K}$ for $c \leq 0.7$. The quadrupolar part of the rate, R_Q , was extracted. It shows essentially the same concentration dependence as existing data for R_Q of Ga in liquid $\text{Ga}_c\text{Te}_{1-c}$, and correlates strongly with the dip in conductivity near $c=0.4$. A quantitative analysis of the concentration dependence is attempted using a simple microscopic model. It comprises localization of conduction electrons including one free parameter for charge shift in the Ga-Te bond (ionicity). Satisfactory agreement is obtained.

Measurements of the nuclear spin relaxation rate R should be very helpful in understanding the metal- semiconductor transition from a microscopic point of view. We report here on measurements of the quadrupolar rate $R_Q(c)$ of As as an impurity atom in liquid $\text{Ga}_c\text{Te}_{1-c}$, which shows a strong correlation with electrical conductivity $\sigma(c)$, as has been observed earlier for R_Q of Ga /1, 2/. Warren /1, 2/ has suggested a connection of the rise in R_Q near $c = 0.4$ with slowing down of the atomic motion and covalent bonding.

We have measured the nuclear spin relaxation rate $R = R_Q + R_M$ in liquid $\text{Ga}_c\text{Te}_{1-c}$, $1 \geq c \geq 0.2$, using $^{73\text{m}}\text{As}$ ($5.6 \mu\text{s}$ state) as the probe nucleus in a TDPAD experiment. Details about the experiment will be given in a subsequent publication /3/. The temperature was 1110 K except for $c = 1$, where $T = 300 \text{ K}$. The magnetic contribution R_M could safely be estimated from the measurement for $^{69,71}\text{Ga}$ /2/. The results for R_Q of $^{73\text{m}}\text{As}$ are presented in the figure. It is noteworthy that they display the same strong concentration dependence as R_Q of Ga /2/, in spite of the considerable chemical difference between Ga and As.

The motional narrowing result for R_Q is $R_Q = \text{const} \cdot \langle V_{20}^2 \rangle \cdot \tau$ where $\langle V_{20}^2 \rangle(c)$ is the mean square fluctuation of the electric field

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gradient at the nucleus (efg), and $\tau(c)$ is the correlation time of V_{20} . Since we know of no other suitable data about atomic dynamics in liquid Ga-Te, R_Q is compared in fig. 1 with viscosity η from /4/. Though this estimate of the concentration dependence due to τ is admittedly very uncertain, we consider it a realistic or even (from liquid metallic alloys) upper estimate for possible changes of $R_Q(c)$ caused by $\tau(c)$. Now $\eta(c=0.4)/\eta(1) \ll R_Q(0.4)/R_Q(1)$, and the temperature dependence of R_Q near $c=0.4$ /3/ is also definitely stronger than that of η . We feel therefore forced to turn to the concentration dependence of $\langle V_{20}^2 \rangle$ and shall attempt a quantitative estimate of the effect of electron localization on it.

For the arrangement of neighbour atoms around the probe a model in \vec{r} -space was used, which reproduces rather well the cancellation of two- and three particle terms in monatomic liquids /3/. The efg produced by a neighbour $i=\text{Ga}$ or Te , V_{20i} is derived from a screened Coulomb potential,

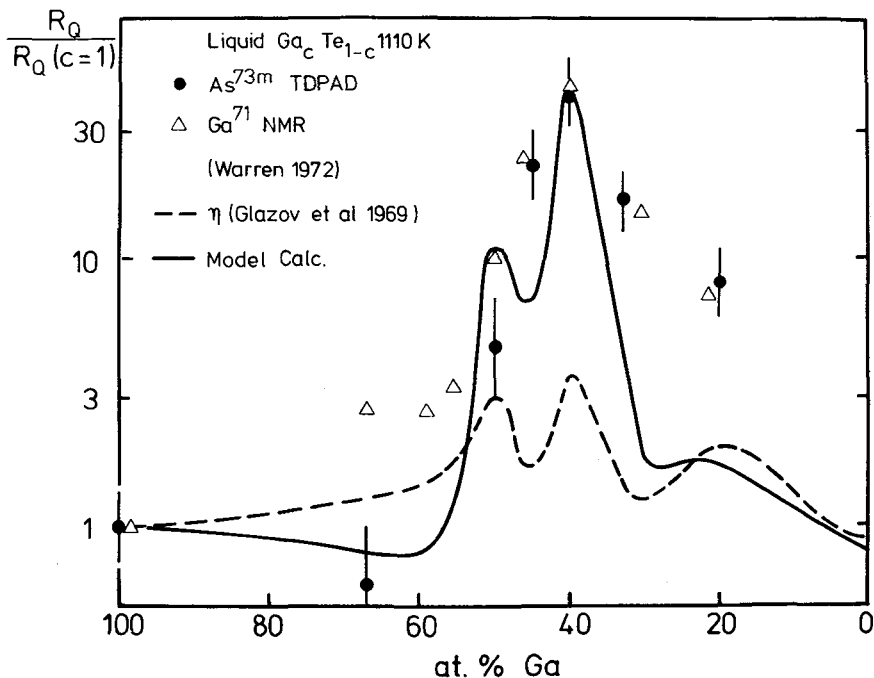


Fig. 1: Concentration dependence of quadrupolar nuclear spin relaxation rate R_Q in liquid $\text{Ga}_c \text{Te}_{1-c}$, normalized at $c=1$. Points are experimental data: △ Warren /2/, ● present work. Full curve is the model calculation described in the text; the free parameter is chosen as $\delta_{\text{ion}}=0.5$. Viscosity η is from /4/.

$$V_i = z_i e r^{-1} e^{-\lambda r}$$

As in an earlier note we thus start from the metallic liquids Ga,Te /5/. As a first approximation, the concentration dependence of the effective Sternheimer factor is neglected. The density of "free" electrons, n^{cond} , determines the conductivity σ , and also the shielding length according to $\lambda^2 = m e^2 n^{-2} (3/\pi \cdot n^{\text{cond}})^{1/3}$. The number of free electrons is 3/atom in Ga. For Te, it is taken from R_{Hall} and σ to be also close to 3 above 1000 K, see /6/. Thus $z_{\text{Ga}}^{\text{ion}} = z_{\text{Te}}^{\text{ion}} = +3$ and $n^{\text{cond}} = 3/v$ in the pure liquids, $v = \text{atomic volume } (v_{\text{Te}} \approx v_{\text{Ga}})$. Comparing $\sigma_{\text{NFE}}(c)$ - obtained by interpolating $1/\sigma_{\text{Ga}}$, $1/\sigma_{\text{Te}}$ - with the experimental $\sigma(c)$ one obtains

$$n^{\text{cond}} = 3/v \cdot \sigma(c) / \sigma_{\text{NFE}}(c) \quad .$$

The average number of localized electrons per cell, $z^{\text{loc}} = 3 - n^{\text{cond}} \cdot v$ is now assigned to Ga or Te cells where their charge adds to z_i^{ion} : $z_i = z_i^{\text{ion}} - z_i^{\text{loc}}$; $z_{\text{Ga}}^{\text{loc}}$ and $z_{\text{Te}}^{\text{loc}}$ can be thought of as composed of a part symmetric with respect to Ga or Te, and an asymmetric part, and the former may be identified with bonding of covalent, the latter with ionic character. The corresponding parameter $\delta_{\text{ion}} (0 \leq \delta_{\text{ion}} \leq 1)$ is normalized such that if at the 2:3 composition full localization occurred ($z^{\text{loc}} = 3$) with 100% ionicity ($\delta_{\text{ion}} = 1$), then the localized electrons would all be found in the Te cells

$$z_{\text{Te}}^{\text{loc}} - z_{\text{Ga}}^{\text{loc}} = (3 + \frac{2}{3} \cdot 3) \cdot z^{\text{loc}} / 3 \cdot \delta_{\text{ion}}$$

This relation and the requirement of charge neutrality, $c z_{\text{Ga}}^{\text{ion}} + (1-c) z_{\text{Te}}^{\text{ion}} = n^{\text{cond}} \cdot v$, fix $z_{\text{Ga}}^{\text{loc}}$ and $z_{\text{Te}}^{\text{loc}}$.

$\langle V_{20}^2 \rangle (c)$ can then be calculated for given c , $\sigma(c)$ and δ_{ion} . It is the difference of charges $z_{\text{Ga}} - z_{\text{Te}} \sim \delta_{\text{ion}}$ which produces a difference in efg's, these being strongly enhanced around $c = 0.4$ by the decrease in λ . A large $\langle V_{20}^2 \rangle$ results from there while in the present treatment the symmetric part of z_i^{loc} contributes little to $\langle V_{20}^2 \rangle$ around $c = 0.4$. (A large difference in efg's may, however, also be ascribed to covalent bonding of the probe atom predominantly to one kind of neighbours.) As discussed above, we use $n(c)$ to estimate $\tau(c)$. The data used are for 1110 K except for the normalization point at $c = 1$ where they are for 300 K.

The full curve in fig. 1 is $R_Q(c)/R_Q(c=1)$ for a value of the single free parameter $\delta_{\text{ion}} = 0.5$ (independent of c). Though there are discrepancies in detail, the overall agreement of the present very

simple model with experiment over the full concentration range is encouraging. The temperature dependence is also satisfactorily explained: for these data and an extended report see /3/.

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