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TITLE: EFFECTS OF SUPERCONDUCTIVITY ON RAPE-EARTH-ION DYNAMICS IN
 $(\text{Ho}_x \text{Lu}_{1-x})\text{Rh}_4\text{B}_4$

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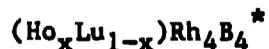
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EFFECTS OF SUPERCONDUCTIVITY ON RARE-EARTH-ION DYNAMICS IN



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ABSTRACT

The temperature dependence of the Ho-ion fluctuation rate τ_m^{-1} in the pseudoternary compound $\text{Ho}_x\text{Lu}_{1-x}\text{Rh}_4\text{B}_4$ is obtained using the zero-field muon spin relaxation technique, primarily for $x = 0.02$. A sharp reduction in the magnitude and a change in the temperature dependence of τ_m^{-1} occur at the superconducting transition temperature T_g . Above T_g the temperature dependence of τ_m^{-1} can be attributed to an activated relaxation process, whereby fluctuations occur by excitation to the first few crystal-field-split states. Neither the temperature dependence nor the sharp change in τ_m^{-1} at $T < T_g$ are presently understood. The data are contrasted with the recent NMR data of Kumagai and Fradin for dilute Gd- and Er-based rhodium borides, which have very different crystal-field-induced level schemes from $\text{Ho}_x\text{Lu}_{1-x}\text{Rh}_4\text{B}_4$.

I. INTRODUCTION

The study of the interplay of magnetism and superconductivity, a topic of interest for some time in solid-state physics,¹ has received renewed interest following the discovery by Matthias and coworkers² of rare-earth (RE) ternary and pseudoternary compounds which exhibit both magnetic and superconducting phase transitions. A particular class of these compounds, the rare-earth rhodium borides (RERh_4B_4), has been extensively studied,³ and thus the ferromagnetic, superconducting, and paramagnetic phase boundaries for these systems are by now well known.^{3,4}

In previous publications^{5,6} we have reported muon spin relaxation (μSR) measurements in the compounds $(\text{Ho}_x\text{Lu}_{1-x})\text{Rh}_4\text{B}_4$ and $(\text{Gd}_x\text{Lu}_{1-x})\text{Rh}_4\text{B}_4$ for $x = 0, 0.7, \text{ and } 1.0$. The μSR technique is an excellent local probe of the rare-earth dynamics in these systems, both because the μ^+ is frozen in the lattice below about 200 K ⁵ and because the measurements can be performed without the perturbative effects of an applied field. A comparison of the μSR measurements in the Ho- and Gd-based compounds establishes that the Ho-ion dynamics are strongly influenced by the crystal-field level splittings. The most striking result, however, is a sharp shoulder in the temperature dependence of the Ho spin-lattice relaxation rate at the superconducting transition temperature $T_g \sim 7.7 \text{ K}$ for $x = 0.7$. In order to investigate further the effects of superconductivity on the Ho-ion dynamics, we have undertaken new measurements for the $x = 0.35$ and 0.02 systems. Here we report primarily zero-field measurements in $(\text{Ho}_{0.02}\text{Lu}_{0.98})\text{Rh}_4\text{B}_4$, which becomes superconducting at $T_g = 11.3 \text{ K}$.

II. EXPERIMENTAL DETAILS

The experiments were performed at the Clinton P. Anderson Meson Physics Facility (LAMPF) using the zero-field, time-differential μ SR technique.⁷ Briefly, a beam of spin-polarized muons is stopped in the sample of interest, and the relaxation rate of the muon spin is monitored by observing the time evolution of the spatial distribution of muon decay positrons relative to the initial muon polarization direction. The sample temperature was varied between 3 K and room temperature and was regulated to within 0.1 K. The sample preparation is described in Ref. 5.

III. DATA ANALYSIS

At different temperatures three distinct forms for the muon spin relaxation function $G_z(t)$ were observed. These are plotted in Fig. 1.

A. $T = 50$ K

At 50 K $G_z(t)$ was found to be well described by the static zero-field relaxation function derived by Kubo and Toyabe^{7,8}. This form of $G_z(t)$ is appropriate for a time-independent, Gaussian distribution of local magnetic fields at the μ^+ site. The measured width of the field distribution, about 4 gauss, indicates that the μ^+ spin relaxation is caused primarily by μ^+ precession about the host nuclear dipolar fields. The return of the long-time portion of $G_z(t)$ to one-third indicates that the local field is effectively random in direction and static in nature.^{7,8} Furthermore, the Ho-ion spins must be fluctuating so rapidly that they produce no relaxation of the μ^+ polarization at this temperature.

B. $15 \text{ K} < T < 50 \text{ K}$

As the temperature is lowered the form of $G_2(t)$ changes, until at 15 K the relaxation function is well-described by a simple exponential (Fig. 1) $G_2(t) = \exp(-\lambda_1 t)$, where λ_1 is the μ^+ spin-lattice relaxation rate. The dominant relaxation mechanism at $T = 15 \text{ K}$ is thus produced by the fluctuating H_0 moments, as discussed below. In the temperature region $15 < T < 50$, the effective $G_2(t)$ was taken to be the product of an exponential and the Kubo-Toyabe function, because the nuclear- and ion-induced broadening mechanisms are independent of each other.

C. $T < T_g$

At $T < T_g$ the form of the relaxation function again changes, exhibiting the two-component form seen in Fig. 1. These data were fit with the function $G_2(t) = (1 - a(T)) \exp(-\sigma t) + a(T) \exp(-\lambda_2 t)$. One finds that:

i) σ is essentially independent of temperature, with an average value of $11.5 \pm 0.6 \mu\text{s}^{-1}$, or about 130 G.

ii) $a(T)$ is about 0.6 at $T = 11 \text{ K}$, and falls to 1/3 for $T < 8 \text{ K}$ (Fig. 2, top).

iii) $\lambda_2(T)$ falls exponentially with decreasing temperature below T_g , with an activation energy $9.8 \pm 1.3 \text{ K}$ (Fig. 2, bottom). From these facts we conclude that the observed two-component relaxation function indicates the onset of quasistatic H_0 -ion motion at and below T_g . The relaxation function is characterized by an inhomogeneous line width σ and a local-field fluctuation rate $\lambda_2(T)$, with amplitude $a(T)$. The 1/3 value of $a(T)$ at low temperatures is indicative of a random, quasistatic field distribution, as for the nuclear case.

We have estimated the expected inhomogeneous line width T_2^{-1} using the formalism of Walstedt and Walker⁹ for dipolar coupling. (The hyperfine coupling of the μ^+ to the conduction electrons is small in metals,¹⁰ and, hence, the μ^+ -ion dipolar coupling dominates indirect mechanisms such as RKKY). To calculate T_2^{-1} one must compute the thermal average of the z-component (perpendicular to the μ^+ spin) of the Ho-ion spin, $\langle S_z \rangle$. As discussed below, the crystal fields produce a two-level, Ising-like wave function for the Ho-ion ground state. This, together with the assumption of short-range ferromagnetic order,¹¹ yields $\langle S_z \rangle = 1/2$. Using $g_{\text{eff}}\mu_B = 10.25 \mu_B$,¹¹ we find $T_2^{-1} = 8 \mu\text{s}^{-1}$, or about 95 G (using the muon gyromagnetic ratio of $8.51 \times 10^4 \text{ s}^{-1}\text{G}^{-1}$). This is in reasonable agreement with the value of σ which we measure, confirming the assignment of dipolar coupling and the association of σ with inhomogeneous broadening.

IV. RARE EARTH DYNAMICS

A. $T > T_g$

At temperatures $T > T_g$ the form of the measured relaxation function $G_z(t)$ (Fig. (1)) indicates that the Ho-spin system is in the fast-fluctuating or motional-narrowing regime, characterized by the relation $\sigma\tau_m \ll 1$, where τ_m is the Ho-ion correlation time. The relation between the muon relaxation rate λ_1 and τ_m in zero field is therefore given¹² by an equation of the form $\lambda_1 = C_D\tau_m$. We have calculated the power-averaged, dipolar coupling C_D for the case of dilute, Ising-like magnetic impurities located at random sites relative to the μ^+ , using both longitudinal and transverse relaxation mechanisms.¹² We obtain $C_D = 5.5 \times 10^{14} \text{ s}^{-2}$. Alternatively, one may estimate the coupling from the inhomogeneous line-width σ , assuming that the μ^+ experiences a sudden

change in the local dipole field resulting from uncorrelated Ho-spin fluctuations (the "strong-collision" model⁷). One finds $C_D = K\sigma^2$, where K is of order 5, and thus $C_D = 6.6 \times 10^{14} \text{s}^{-2}$, in good agreement with the other method of calculation. The values of τ_m^{-1} obtained in this way are shown in Fig. 2 for $T > T_g$.

B. $T < T_g$

In the region of quasistatic Ho-ion motion, the μ^+ spin follows the changing local-dipolar field adiabatically. Specifically, one has⁷ $\tau_m^{-1} = 1.5 \lambda_2$. The temperature dependence of τ_m^{-1} for $T < T_g$ is shown in Fig. 2.

V. DISCUSSION

The cause of Ho-ion relaxation is of considerable interest. Two basic mechanisms are possible, indirect spin-spin exchange coupling (RKKY)¹² or direct relaxation due to s-f exchange (Korringa mechanism).¹³ For the RKKY process, τ_m^{-1} is proportional to x , while τ_m^{-1} is independent of x for the Korringa mechanism. The dipole coupling strength C_D is proportional to x^2 . At $T = 100$ K, the measured μ SR rate λ_1 for $x > 0.35$ is proportional to x^2 (Fig. 3), indicating that the Korringa process is dominant. The same mechanism has also been observed in SmRh_4B_4 at high temperatures.¹⁴ For $11 \text{ K} < T < 50 \text{ K}$ the μ SR rate is too large to be observed,⁵ and hence the concentration dependence of τ_m^{-1} cannot be determined. Nevertheless, it is still possible to infer the relaxation mechanism.

For very dilute Gd- and Er-based systems, Kumagai and Fradin¹⁵ report that the RKKY process is dominant for $T = T_g$. The low-lying eigenstates of the crystal-field Hamiltonian are very different in these systems compared to $(\text{Ho}_x\text{Lu}_{1-x})\text{Rh}_4\text{B}_4$, however. We have computed the $(\text{RE})\text{Rh}_4\text{B}_4$ level

structure by diagonalizing the Hamiltonian for $\bar{4}2m$ point symmetry using the crystal-field parameters of Dunlap and Niarchos.¹⁶ In the Ho systems the ground and first-excited states are each doubly degenerate, with nearly pure $|±8\rangle$ and $|±7\rangle$ wave functions, respectively. The first excited states occur at $T = 55$ K. Allowed transitions from $|±8\rangle$ to $|±7\rangle$ can therefore only occur through small admixtures of different J_z eigenfunctions in the ground and excited states, which are separated by at least 55 K. Thus, whether the relaxation occurs through Korringa or RKKY processes, the magnitude of τ_m^{-1} in the Ho-based compounds at low temperatures should be significantly smaller than for the Gd or Er systems, where the eigenstates allow significant ground-state relaxation. (The s-f exchange is assumed to be nearly independent of the RE in $RERh_4B_4$ compounds.^{15,17}) In fact, τ_m^{-1} for $(Ho_{.02}Lu_{.98})Rh_4B_4$ at $T = 15$ K is about 25 times greater than for $(Er_{.0002}Y_{.9998})Rh_4B_4$ at the same temperature. We thus infer that the concentration-dependent RKKY process is dominant at $T \approx T_g$ for both compounds. The temperature dependence of τ_m^{-1} in $(Ho_{.02}Lu_{.98})Rh_4B_4$ at $T \gtrsim T_g$ is consistent with an activated process where relaxation occurs only via excited states. A similar behavior was observed in $HoRh_4B_4$.⁵

At $T = T_g$, τ_m^{-1} drops sharply to a value about 100 times smaller than the rate extrapolated from the data at $T > T_g$ (Fig. 2, bottom). In addition, the temperature dependence changes as well. A less drastic change in RE dynamics for $T < T_g$ is found by Kumagai and Fradin for RE = Gd, Er systems,¹⁵ albeit in an applied magnetic field. How this large change in Ho dynamics occurs is unknown, but the solution is likely tied to a description of the exchange mechanism in the presence of both superconductivity and the low-lying, crystal-field states. In particular, the inverse-temperature dependence of τ_m^{-1} appears to be exponential

(though measured only over 1 decade), but with an activation energy about 1/5 of the ground to first-excited-state splitting. NMR data in YRh_4B_4 ¹⁵ at $T < T_g$ are consistent with a superconducting gap parameter $2\Delta_g = 6.6 T_g$, in contrast to the usual BCS value of $3.5 T_g$. Interpreted in this way the exponential activation energy which we measure corresponds $2\Delta_g = 1.8 T_g$. Such an association is speculative at this point, however. Finally, the gradual change of the fluctuating amplitude $a(T)$ for $T < T_g$ (Fig. 2, top) may be due to the presence of a mixed superconducting state, which only becomes homogeneous as the temperature is lowered well below T_g .

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FIGURE CAPTIONS

1. Zero-field muon spin relaxation function $G_z(t)$ vs. the time between μ^+ stop and decay. The lines are fits to the respective functions as discussed in the text.
2. Top: Amplitude of the fluctuating component of the muon spin relaxation function $G_z(t)$ vs. temperature.
Bottom: Ho-ion fluctuation rate τ_m^{-1} vs. inverse temperature. Note the break in logarithmic scale. The dotted lines in both top and bottom are shown to guide the eye.
3. Muon spin relaxation rate λ_1 vs. square of Ho-ion concentration in $\text{Ho}_x\text{Lu}_{1-x}\text{Rh}_4\text{B}_4$. Data for $x = 1.0, 0.70, 0.35,$ and 0.02 are shown. The $x = 0.02$ datum is at the origin.

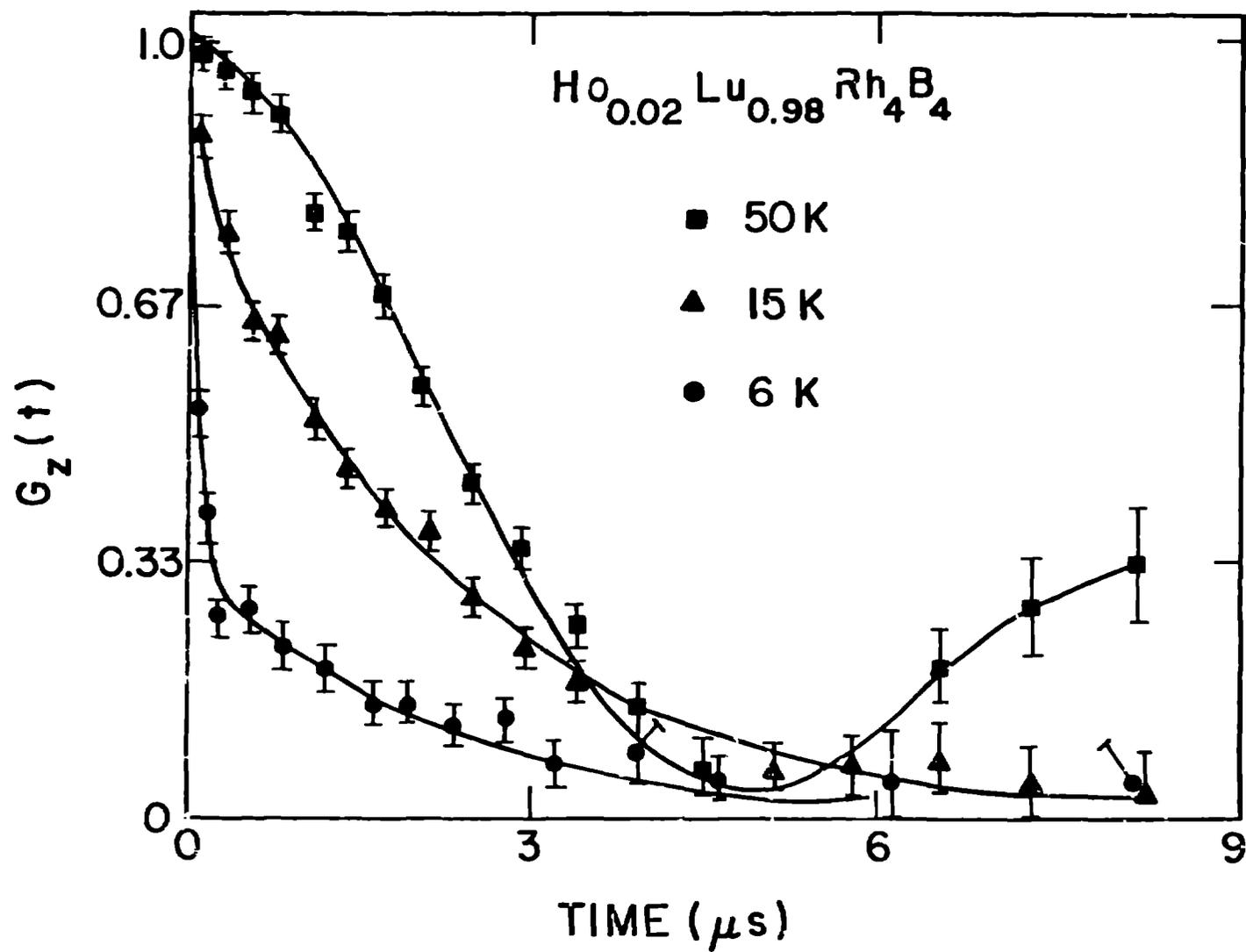


Fig 1



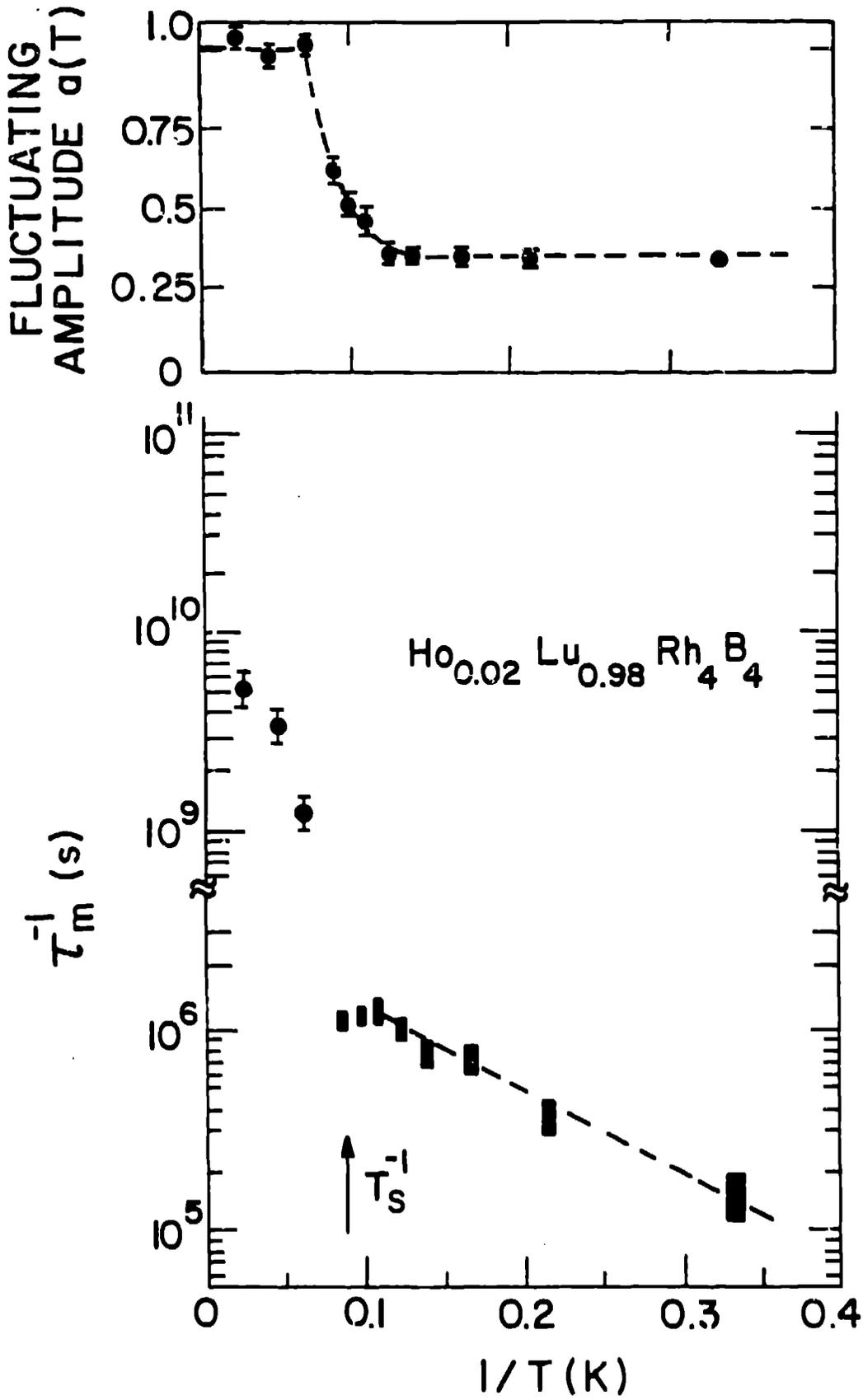


Fig 2

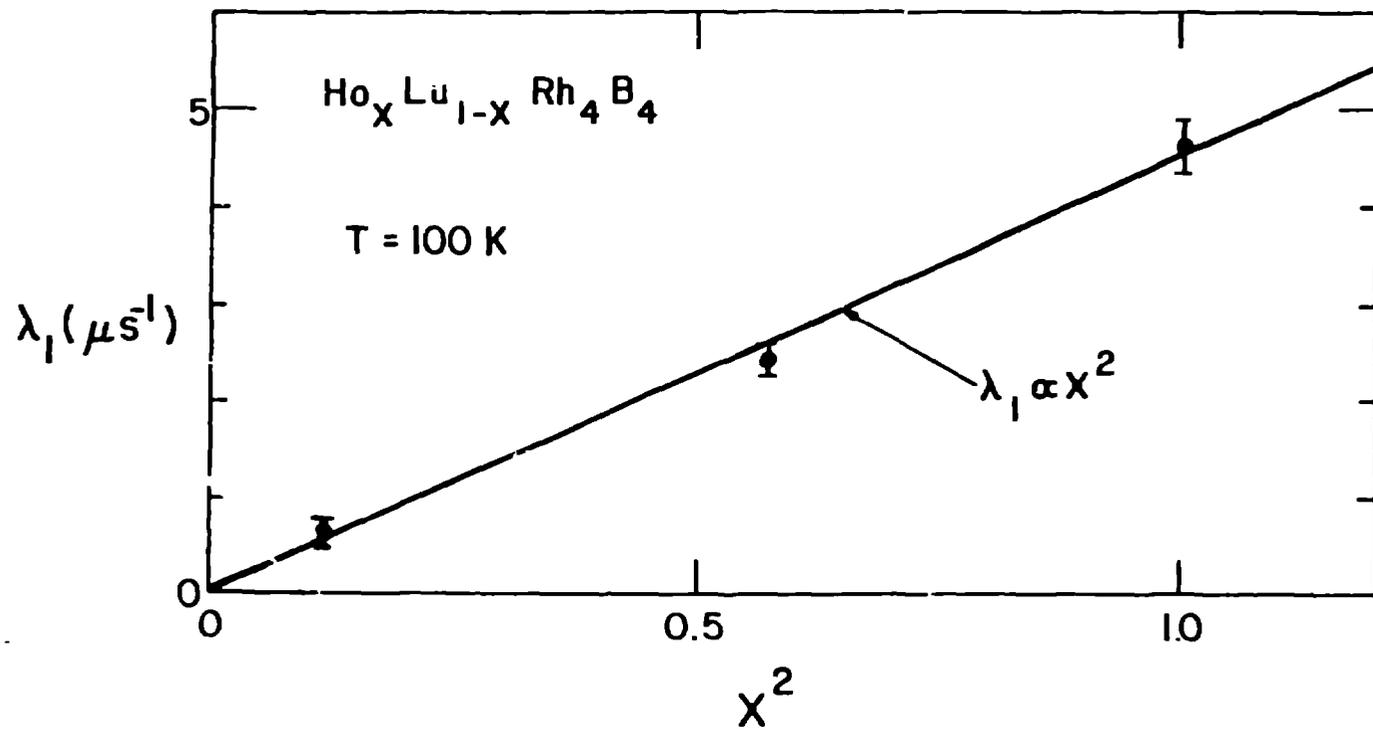


Fig 3