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μ^+SR Spectroscopy: The Positive Muon as a Magnetic Probe in Solids

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ABSTRACT

Through its asymmetric decay, the positive muon acts as a sensitive detector of the interactions of its spin with the medium in which it comes to rest. Development of the μ^+SR spectroscopy technique is described, and recent applications of the μ^+ as a probe are discussed. Results for hyperfine fields in ferromagnets and impurity states in nonmetals are presented with suggestions for future studies.

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μ^+SR Spectroscopy: The Positive Muon as a Magnetic Probe in Solids

INTRODUCTION TO μSR

Muons are created and destroyed in the maximally parity-violating decays $\pi \rightarrow \mu + \nu$ and $\mu \rightarrow e + \nu + \bar{\nu}$. The former reaction produces muons of definite helicity, which can be collected into beams which are nearly 100% polarized.[1] In the latter reaction the $e^{\pm}$ is preferentially emitted along (for μ^+) or opposite to (for μ^-) the muon spin, in a pattern of the form $D(\theta) \sim 1 \pm a \cos\theta$, where θ is the angle between $\mu^{\pm}$ spin and $e^{\pm}$ direction, and a is an asymmetry parameter -- a fuction of $e^{\pm}$ energy -- whose average over energy is 1/3.[2] The weak interaction thus provides experimenters with a source of highly polarized Dirac particles and a convenient way to detect their polarization. Such structureless, singly charged particles are especially attractive as probes of matter.[3]

A typical experimental technique[4] is to stop a beam of polarized muons in a target, starting a "clock" as a muon enters through scintillation counters, and stopping the "clock" when a decay electron (positron) is counted in a second scintillator array. When a magnetic field is applied transverse to the muon polarization, the spin of the muon will precess at its Larmor frequency, causing the $e^{\pm}$ detection probability to become an oscillatory function of time as the muon spin sweeps past the $e^{\pm}$ telescope. The histogram of measured times, an example of which is shown in Fig. 1, is generally fitted to the form

$$N(t) = N_o \{e^{-t/\tau_\mu} \left(1 + A\, e^{-t/T_2} \cos\, (\omega_\mu t + \phi)\right) + B\}, \qquad (1)$$

where N_o is a normalization factor, τ_μ is the free muon lifetime (2.2 μsec for μ^+ or free μ^-), A is the apparent initial asymmetry, T_2 is a transverse relaxation time (accounting for slow depolarization by random local fields), ω_μ is the muon Larmor frequency, ϕ is the apparent initial phase of the precession, and B is a constant background from random events.

This technique, which is clearly analogous to nuclear time differential perturbed angular distribution (TDPAD) methods, has been dubbed "μSR" (for Muon Spin Rotation) in an attempt to suggest the many applications analogous to those of NMR and ESR.[3] Each of the fitted parameters in Eq. 1 is of special interest in certain studies: for μ^-, $1/\tau_\mu$ is a measure of the combined rates of decay and capture in various nuclei, providing information on weak processes[5] as well as nuclear structure;[6] the asymmetry A and the phase ϕ determine the magnitude and direction of the apparent initial muon polarization, variations in which can be interpreted in terms of chemical processes involving muons;[7,8] the precession frequency ω_μ and the relaxation rate $1/T_2$ are most interesting for the study of solids, since they give a direct quantitative measure of the local field B_μ at the site of the muon, and its inhomogeneity ΔB_μ:

$$\omega_\mu = \gamma_\mu B_\mu \quad \text{and} \quad \frac{1}{T_2} = \tfrac{1}{2}\, \gamma_\mu\, \Delta B_\mu, \qquad (2)$$

where $\gamma_\mu = 0.85 \times 10^5$ rad/sec-G (period = 74 nsec at 1 kG).

In many solids, distinct fractions of the muon ensemble may see entirely different values of B_μ and ΔB_μ; in such cases, or in any case when B_μ is not even approximately known ahead of time, the

"μSR spectroscopy" technique is especially useful: the constant background is subtracted and the exponential μ decay divided out of N(t), which is then Fourier transformed to produce a frequency spectrum, in which a "signal" of width $1/T_2$ at a frequency ω_μ reflects the local field $B_\mu \pm \Delta B_\mu$, just as in NMR. An example of such a spectrum is shown in Fig. 2.

Conventional μ^+SR methods with positive muons have opened up a number of new fields to investigation;[3] μ^-SR studies with negative muons are also of growing interest.[8,9] Here we wish to concentrate on several of the more dramatic results which have recently appeared through the new technique of μ^+SR spectroscopy.

I. INTERSTITIAL HYPERFINE FIELD IN FERROMAGNETS

A. μ^+SR in Magnetic Media

Once thermalized in condensed matter, the positive muon behaves as if it were a light-proton ($m_p/m_\mu = 8.9$); thus in most crystals it can be expected to take up an interstitial position. It then experiences a local field which has the following contributions:

$$B_\mu = B_{ext} + B_{dm} + B_L + B_{dip} + B_{hf}, \qquad (3)$$

where B_{ext} is the applied field, B_{dm} is the sample demagnetizing field, B_L is the field due to induced magnetic charges on an imaginary spherical "Lorentz cavity" centered about the μ^+ site, B_{dip} is the field due to the local dipole moments within the Lorentz sphere, and B_{hf} is a contribution due to contact interactions with polarized electrons.

In a ferromagnetic crystal below saturation, B_{ext} is just cancelled

by B_{dm}. B_L has the value $\frac{4\pi}{3} M_s$, where M_s is the saturation magnetization in a domain (the imaginary Lorentz cavity must be kept smaller than the dimensions of one domain). B_{dip} is usually calculable if the favored interstitial site can be identified. We can then obtain a value for B_{hf}, a quantity of basic interest to the physics of ferromagnetism. The advantage of the μ^+ for such a measurement is the lack of any complications related to an electronic core.

B. μ^+SR in Nickel

Measurements of ω_μ and T_2 for positive muons in ferromagnetic Ni have been made by several groups[10,11] over a range of temperatures, yielding the data shown in Fig. 3 for B_μ and ΔB_μ. The temperature dependence is essentially a Brillouin function with the same shape as the saturation magnetization curve, normalized to a zero-temperature limit of $B_\mu(0) = + 1480$ G.

In Ni, the μ^+ can be expected to occupy an octahedral interstitial site with cubic symmetry. The local dipole contributions then cancel, giving $B_{dip} = 0$. The Lorentz cavity contribution is just $B_L(0) = + 2140$ G, leaving a hyperfine contribution of $B_{hf} = - 660$ G. This field is thought to be due to the contact interaction between the μ^+ and polarized 4s-band conduction electrons.

Neutron diffraction studies[12] have indicated an unperturbed average interstitial electron polarization of - 17 %, implying a local interstitial magnetization of $M_{loc} = - 79$ G. An uncharged probe would thus measure a contact field of $B_{hf} = \frac{8\pi}{3} M_{loc} = - 660$ G. The agreement between this value and the measured field must be fortuitous, since the μ^+ charge

must be screened. If we assume that only 4s-band electrons (treated as a Fermi gas of uniform density) contribute to the screening, the Fermi-Thomas screening length is ~ 0.6 Å; using the Linhard dielectric function approach, we estimate a concentration of electron density at the μ^+ by a factor of ~ 5 over the unperturbed interstitial electron density. If the screening electrons had the same polarization as the band from which they come, one would expect a contact field of $B_{hf} = -3300$ G. To explain the much lower measured value we are studying a simple model in which local energy density minimization favors equalized screening contributions from spin up and spin down electrons.[13]

C. μ^+SR in Iron

Unlike Ni, Fe has two electrostatically identical but magnetically different interstitial face-centered sites. One has a B_{dip} contribution of + 18,800 G, while the other has $B_{dip} = -9400$ G and occurs twice as often. Thus at high temperatures the μ^+ diffuses between sites fast enough for B_{dip} to average to zero, but at low temperature this "motional narrowing" effect is absent and the μ^+ is rapidly depolarized.[14] Preliminary attempts to "freeze" muons into distinct sites in a single crystal of Fe at 77°K have so far yielded inconclusive results.

High temperature data for B_μ vs. T can be extrapolated to T = 0 to give a value of $B_\mu(0) = +4100$ G. Using $\langle B_{dip}\rangle = 0$ and $B_L(0) = \frac{4\pi}{3} M_s(0) = +7100$ G in Eq. (3) gives $B_{hf}(0) = -3000$ G. Interpretation of this result is complicated by the highly structured spatial distribution of the interstitial magnetization in iron,[15] since the zero-point motion of the μ^+ in the interstitial potential well may be significant.

D. Vibration and Diffusion of the μ^+

Interpretation of μ^+SR results is often plagued by two uncertainties which we have mentioned in the case of iron. First, how localized is the μ^+ within its interstitial site? And second, how rapidly does it "jump" from site to site? The first question relates to the zero-point vibrational motion, which can presumably be estimated as follows: if we assume a simple harmonic oscillator potential for the interstitial "well," the amplitude of the zero-point motion scales as $m^{-1/4}$. Thus the zero-point motion of a μ^+ should have an amplitude about 1.71 times as large as that of a proton in the same site. Information on such motions of protons can be gathered by quasielastic neutron scattering.[16]

The second question is one of the diffusion rate -- or equivalently of the height and width of the potential barrier between sites, which the μ^+ must either tunnel through or be thermally excited over in order to diffuse. Again there is a direct analogy with the study of diffusion of hydrogen isotopes.[16] Studies of the "motional narrowing" of μ^+ relaxation by nuclear moments in copper[17] and ice[18] have shown that in these substances the muon is "frozen" into a single site for its entire lifetime at temperatures below about 80°K. However, in other solids the shape and height of the barrier may be quite different, perhaps even leading to a delocalized μ^+ wave function due to tunneling in some cases.[19] This problem should often be amenable to study by the "motional narrowing" method, and the question of the localization of the μ^+ can and should be answered experimentally.

II. IMPURITY STATES IN INSULATORS AND SEMICONDUCTORS

A. The μ^+ as an Impurity

The μ^+, with its family resemblance to a proton, is perhaps the simplest impurity nucleus one can implant in any crystal. There has been some question in the past as to whether the implanted μ^+ forms substitutional or interstitial states, and whether it remains a bare μ^+, captures an electron to form a muonium (Mu) atom, or perhaps captures two electrons to form a Mu^- ion. It has generally been agreed[20] that a significant fraction must form Mu atoms, but it has not been clear whether these atoms would be "shallow donors," with their electron wave functions spread over many lattice sites, or "deep donors," with the entire atoms fitting into single sites. The time scale for stability of such atoms has also been in question.[21] Recent experimental results using μ^+SR spectroscopy have partially resolved these issues.

B. The Muonium Atom

In a free Mu atom in its ground state, the μ^+ and e^- spins are coupled by a contact interaction of strength $\Delta E_{hf}/h = \nu_0 = 4463$ MHz,[22] given by the splitting between singlet and triplet total angular momentum states:[23]

$$\Delta E_{hf} = \frac{8}{3r_0^3} \; g_e \; \mu_0^e \; g_\mu \; \mu_0^\mu, \qquad (4)$$

where r_0 is the mean radius of the atom, $g_{e,\mu}$ are electron and muon g-factors, and $\mu_0^{e,\mu}$ are electron and muon magnetons. One can think of ΔE_{hf} as the energy of interaction of the electron's magnetic moment with the "contact field" due to the muon, $B_0 = 1593$ G.

In weak applied fields ($B \ll B_0$), the triplet state of Mu will precess at a frequency ω_{Mu}. Since Mu ($F = 1$) has essentially the magnetic moment of an electron, but has twice its angular momentum, ω_{Mu} is about ½ of ω_e, the electron Larmor frequency -- or about 103 times ω_μ, the free μ^+ Larmor frequency. This muonium precession can be observed directly by the μSR method[24,25,26] in fields up to ∿ 200 G.

In stronger fields ($B \sim B_0$), and in all fields to some extent, competition between the Zeeman couplings and the hyperfine coupling causes ω_{Mu} to split into two frequencies, equivalent to the transition frequencies ω_{12} and ω_{23} (where $\omega_{ij} \equiv \omega_i - \omega_j$) between energy eigenstates described by the Breit-Rabi formulae:

$$\omega_1 = \frac{\omega_0}{4} + \omega_-, \qquad \omega_2 = -\frac{\omega_0}{4} + \sqrt{\frac{\omega_0{}^2}{4} + \omega_+^2}\,,$$
$$\omega_3 = \frac{\omega_0}{4} - \omega_-, \qquad \omega_4 = -\frac{\omega_0}{4} - \sqrt{\frac{\omega_0{}^2}{4} + \omega_+^2}\,, \tag{5}$$

where $\omega_\pm = \frac{1}{2}(\omega_e \pm \omega_\mu)$ and $\omega_0 = 2\pi\,\nu_0$. In low fields, the splitting between ω_{12} and ω_{23} is given by

$$\Omega = \tfrac{1}{2}(\omega_{23} - \omega_{12}) \simeq \omega_-{}^2/\omega_0. \tag{6}$$

C. Muonium in Insulators

The splitting of Mu precession into two frequencies leads to "beats" in the μ^+SR precession pattern -- which, along with the high frequency of Mu precession relative to free μ^+ precession, distinctively characterizes the formation of long-lived deep donor Mu atoms. This "two-frequency muonium precession" has been observed in a variety of insulators, including quartz,[24,25,26] ice,[25] and solid CO_2.[27] In all these media, the measured

"beat frequency" Ω is consistent with that predicted by Eq. (6), using the vacuum value for ω_0.

Relaxation of the Mu precession signal in these insulators is apparently due to random local fields from magnetic nuclei, as borne out in a study of muonium relaxation in H_2O and D_2O.[18] In the alkali halides, an extremely fast relaxation of muonium[28] renders Mu precession unobservable; it has been suggested[29] that this relaxation is due to a "superhyperfine" coupling between muon, electron, and nuclear spins. Such a coupling could lead to new coherent precession frequencies; this phenomenon has not yet been observed, but new high-resolution μ^+SR spectroscopy facilities at "meson factories" such as TRIUMF and SIN may facilitate its detection.

D. Deep-donor Muonium in Semiconductors

Observations of two-frequency precession of Mu in pure, cold (77°K) single crystals of germanium[25] and silicon[26] have proven that positive muons stopped in semiconductors also form long-lived, deep-donor interstitial Mu atoms under some circumstances. Fig. 4 shows μ^+SR spectra for SiO_2 and Si in the same field. The peak near zero in each spectrum is due to free μ^+ Larmor precession; the rightmost pair of peaks represent two-frequency precession of deep-donor Mu. The splitting is visibly larger in Si than in SiO_2, where $\nu_0(SiO_2) \approx \nu_0(\text{vac})$. This splitting frequency (2Ω) was measured and used with Eq. (6) to extract the hyperfine frequency for deep-donor Mu in silicon.[26] A similar analysis was made for deep-donor Mu in germanium.[25] The results are

$$\nu_0(\text{Ge})/\nu_0(\text{vac}) = 0.56 \pm 0.01; \quad \nu_0(\text{Si})/\nu_0(\text{vac}) = 0.45 \pm 0.02.$$

Recalling Eq. (4), we see that the radii of Mu atoms in these crystals are about 1.2 times larger than in vacuum; thus they are clearly deep-donor impurities. The "swelling" of the atom is due to screening by valence band electrons from neighboring lattice sites; its magnitude gives new information about the crystals.[30]

E. Shallow-donor Muonium in Semiconductors

Also evident in the Si spectrum are two "anomalous" peaks at intermediate frequencies. At first these signals caused great confusion, but studies of their field dependence[26] (shown in Fig. 5) revealed that they correspond to transitions ω_{12} and ω_{34} between Breit-Rabi energy levels of a modified Mu atom in which both ν_0 and g_e are quite different from their vacuum values. It was also found that ν_0 was anisotropic, as indicated in Fig. 5. The fitted values were: for the (111) crystal axis along the field, $\nu_0/\nu_0(\text{vac}) = 0.0198 \pm 0.0002$; for the (100) axis along the field, $\nu_0/\nu_0(\text{vac}) = 0.0205 \pm 0.0003$; in both cases, $g_e = 13 \pm 3$.

The preferred interpretation of these results is that the two "anomalous" frequencies are due to precession of shallow-donor Mu atoms, in which the electron wave functions must be superpositions of conduction band states, which may have $\ell \neq 0$ and small anisotropic effective masses.

F. Quasi-free μ^+ in Semiconductors

Mu precession has only been observed in mildly p-type Si and Ge at low temperature; however, in all semiconductor crystals there appears a precession signal of some strength at the free μ^+ Larmor frequency. Whether this signal is due to substitutional μ^+, Mu^- (where the two electrons pair up to make a diamagnetic system), or some diamagnetic

bound state of a μ^+ with lattice defects (perhaps created in its stopping) is not known. However, the μ^+ precession signal in p-type Si at 77°K and 4400 G appears to have two components: one long-lived (the same as is usually observed) and one which dies out within ∿ 30 nsec. These two components must be distinct products of the thermalization process, as explained below.

G. Population of States in Semiconductors

All the precession signals mentioned above are present simultaneously in the Si spectrum shown in Fig. 4. Since these motions are out of phase with each other within a few nsec after the muons stop in the target, they must each represent an independent and isolated long-lived product of the thermalization process, as indicated graphically in Fig. 6.

The amplitudes of the various precession signals are proportional to the respective fractions h, f_+, f_{sd}, and f_{dd}, which should sum to unity but are all strong functions of doping and temperature (see Ref. 21). Studies of their relative populations under different conditions may reveal much about the formation and behavior of hydrogen-like impurity states in semiconductors.[3]

SUMMARY

We have briefly reviewed several applications of the μ^+SR spectroscopy technique to the study of interstitial ions and atomic impurity states in solids. In each case, recent results have greatly clarified the behavior of positive muons in the relevant crystals. However, there are

still many unanswered questions even in these cases; further studies are needed before the rewards in terms of improved understanding of related solid state phenomena begin to be realized. The immediate future of the μSR technique as a tool for the study of such basic fields as hydrogen in metals[16] will be largely determined by the care and depth with which the next round of experiments on these systems are performed.

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FIGURE CAPTIONS

Fig. 1. Typical experimental time histogram, showing μ^+ precession in a CCl_4 target at 100 G.

Fig. 2. Frequency spectrum for μ^+SR in a single crystal of Ni at 20^oK with a 150 G external field along the (111) crystal axis. The vertical scale is the square of the Fourier transform amplitude. The leftmost peaks in the spectrum are due to arbitrary data cutoff at 2.9 μsec; the peak at about 2 MHz is from muons stopped in the cryostat walls, etc., which precess in the external field; the peak at 19.2 MHz is a characteristic "noise" peak due to the rf structure of the 184 in. Cyclotron beam, which "leaks" into the data through the supposedly time-independent accidental background. The peak at 20.05 MHz is from μ^+ precession in the internal field B_μ in Ni.

Fig. 3. Local field B_μ at the muon site and linewidth ΔB observed in Ni by various techniques. The solid curve is the saturation magnetization curve for Ni, normalized to $B_\mu(0) = 1.5$ kG.

Fig. 4. μ^+SR frequency spectra in fused SiO_2 at room temperature and in mildly p-type single crystal Si at 77^oK. Applied field is 100 G in both cases.

Fig. 5. Dependence of "anomalous" frequencies in Si upon field strength and crystal orientation. Round points and solid lines are for (111) crystal axis along the field; triangles and dashed lines are for (100) along the field. Square points (for strong signals) and horizontal bars (for weak signals) indicate unexplained additional peaks.

Fig. 6. Hypothetical model for initial population of states in pure silicon crystals at low temperature.

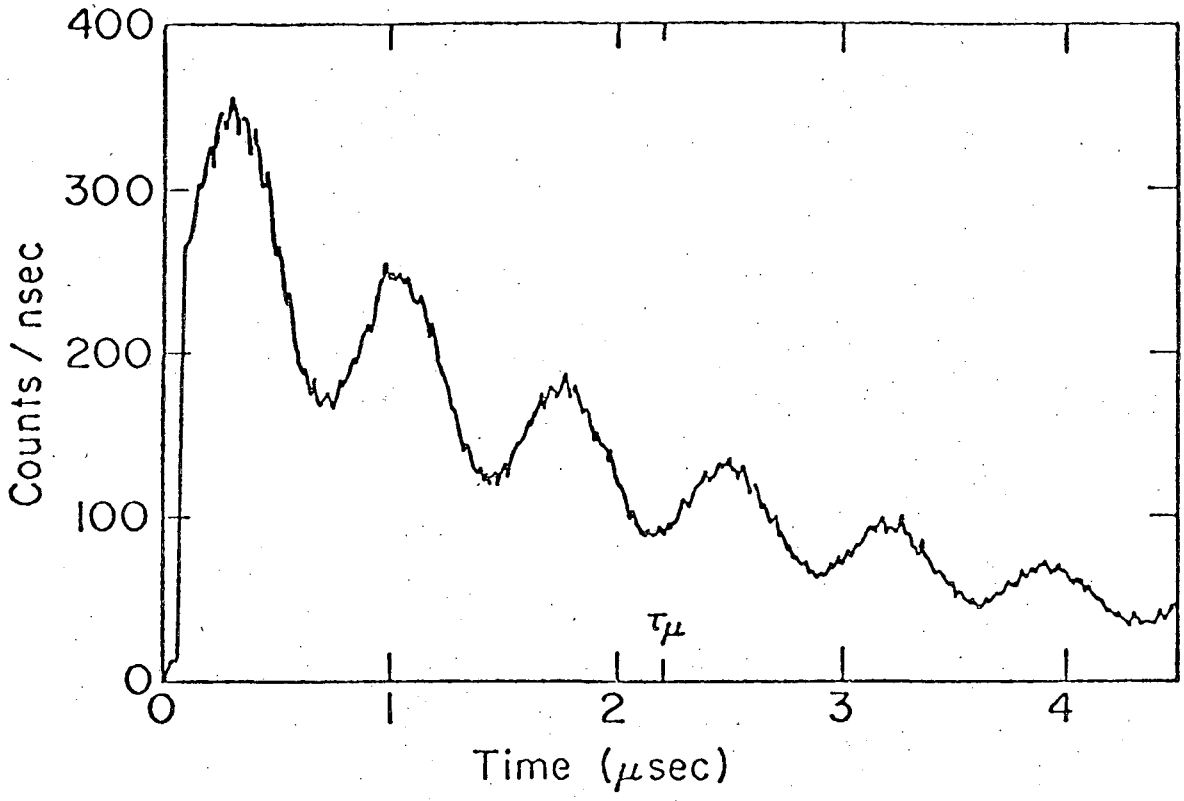

Fig. 1

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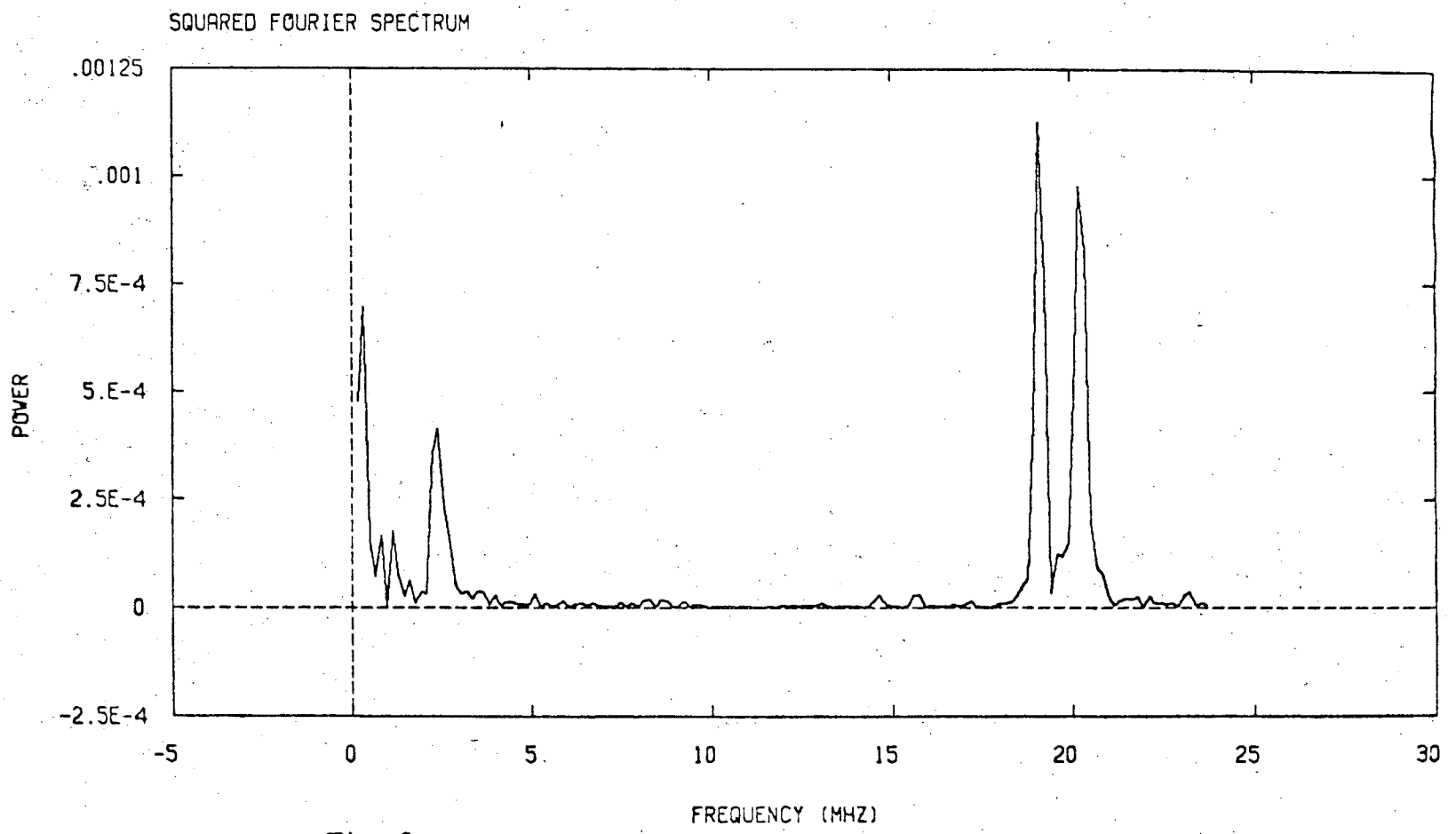

Fig. 2

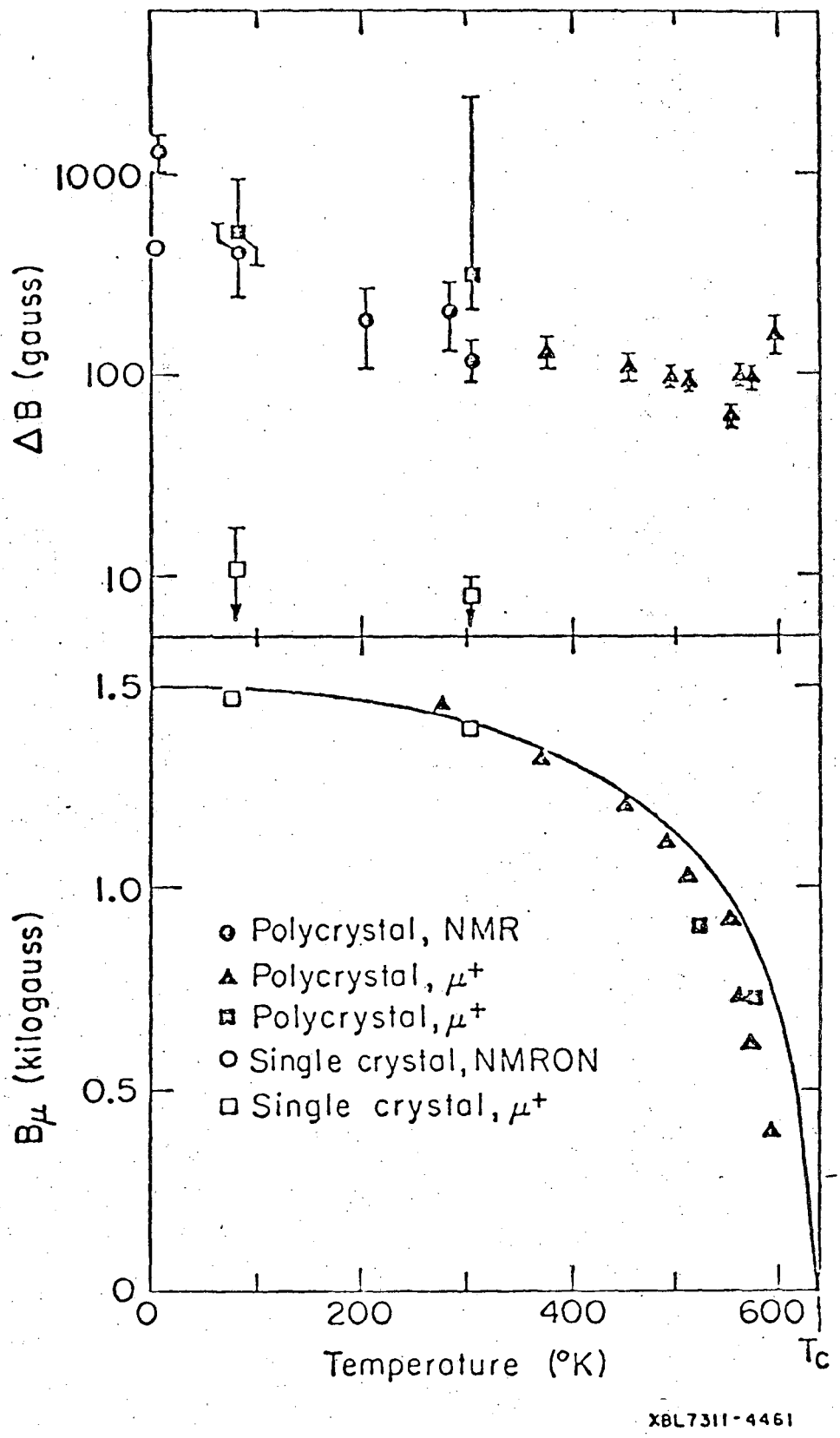
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Fig. 3

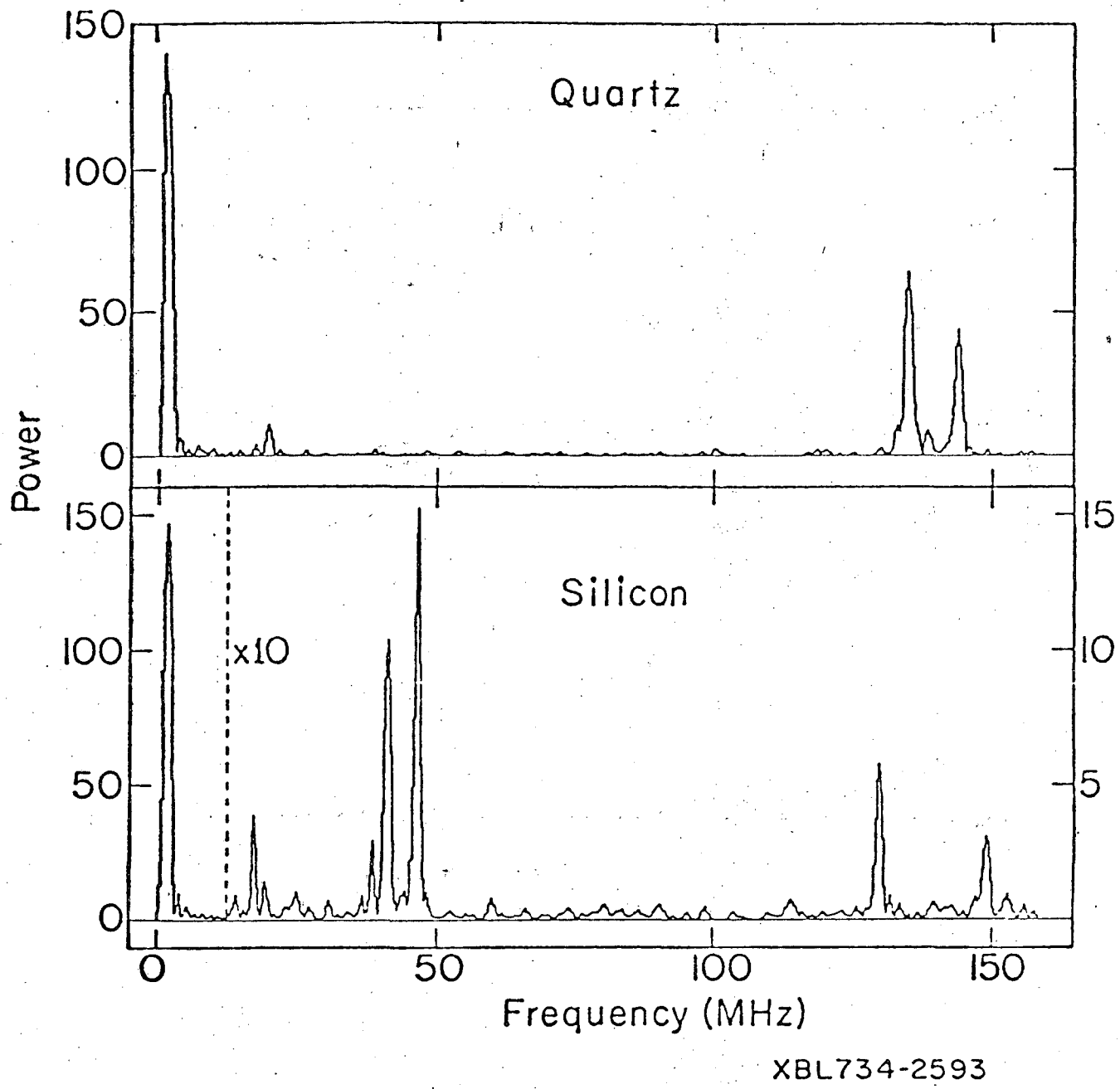
Quartz
Silicon
x10
Power
Frequency (MHz)
150
100
50
0
15
10
5
0
50
100
150
XBL734-2593

Fig. 4

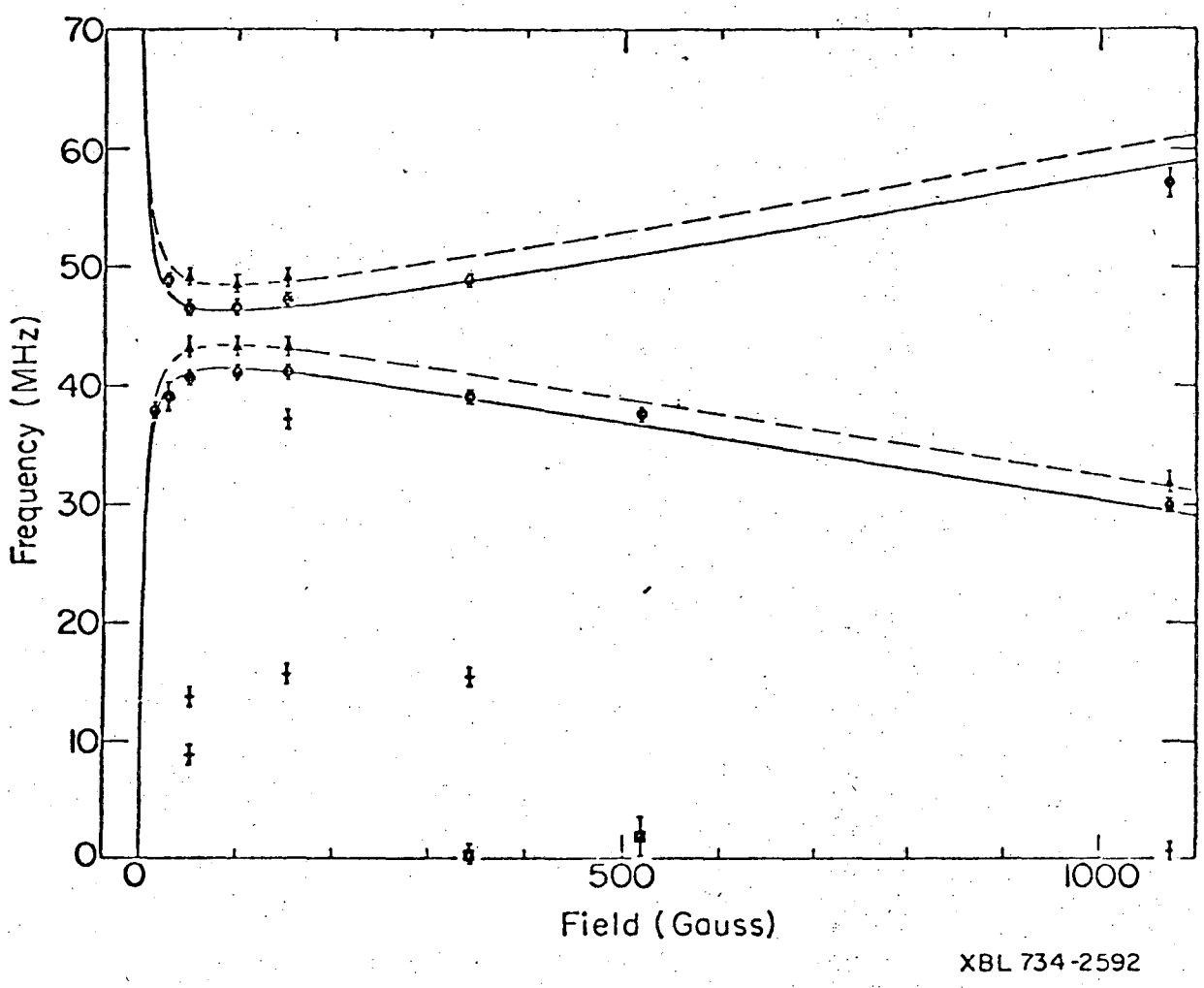
70
60
50
40
30
20
10
0
Frequency (MHz)
0
500
1000
Field (Gauss)
XBL 734-2592

Fig. 5

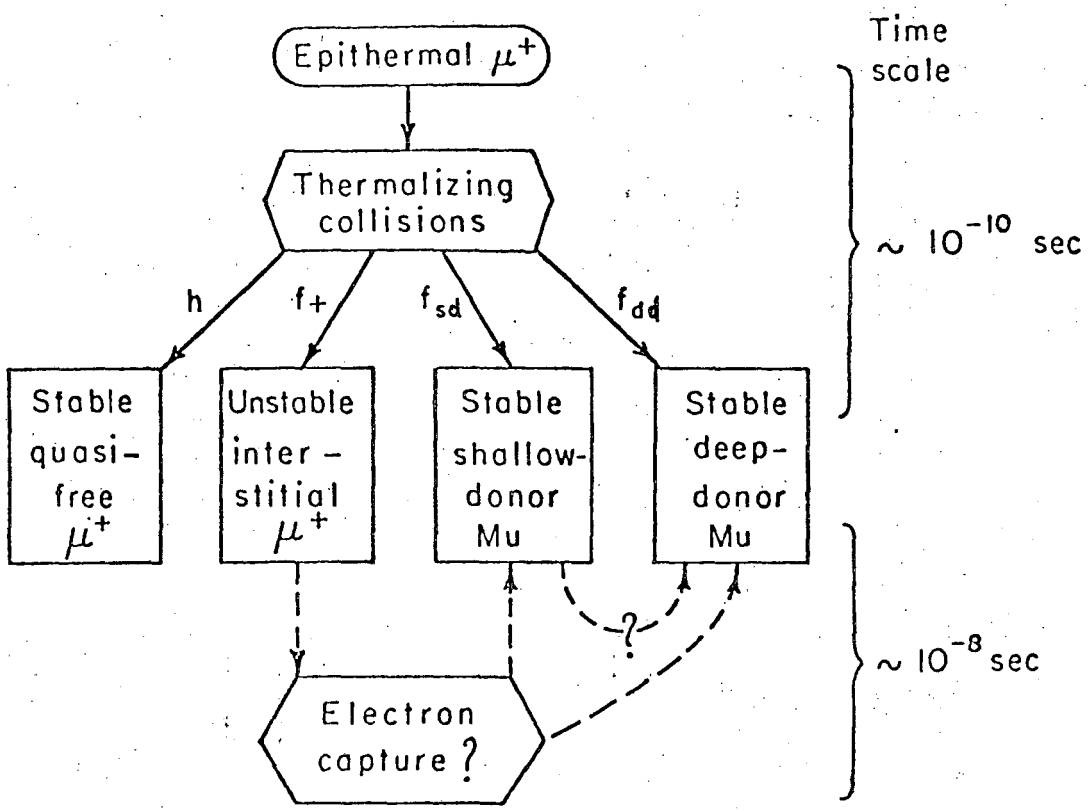
Epithermal μ^+
Thermalizing collisions
h
f_+
f_{sd}
f_{dd}
Stable quasi-free μ^+
Unstable inter-stitial μ^+
Stable shallow-donor Mu
Stable deep-donor Mu
?
Electron capture ?
Time scale
$\sim 10^{-10}$ sec
$\sim 10^{-8}$ sec

Fig. 6

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