STUDIES OF SUPERCONDUCTING MATERIALS
WITH MUON SPIN ROTATION

M.R. Davis and C.E. Stronach
Physics Department
Virginia State University
Petersburg, VA

W.J. Kossler, H.E. Schone, and X.H. Yu
Physics Department
College of William and Mary
Williamsburg, VA

Y.J. Uemura and B.J. Sternlieb
Physics Department
Columbia University
New York, NY

J.R. Kempton
TRIUMF
4004 Wesbrook Mall
Vancouver, BC
Canada

J. Oostens
Physics Department
University of Cincinnati
Cincinnati, OH

and

W.F. Lankford
Physics Department
George Mason University
Fairfax, VA
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ABSTRACT

The muon spin rotation/relaxation technique has been found to be an exceptionally effective means of measuring the magnetic properties of superconductors, including the new high temperature superconductor materials, at the microscopic level. The technique directly measures the magnetic penetration depth (type II SC's) and detects the presence of magnetic ordering (antiferromagnetism or spin-glass ordering has been observed in some HTSC's and in many closely related compounds). Extensive studies of HTSC materials have been conducted by the Virginia State University - College of William and Mary - Columbia University collaboration at Brookhaven National Laboratory and TRIUMF (Vancouver). A survey of LaSrCuO, YBaCaCuO systems shows an essentially linear relationship between the transition temperature $T_c$ and the relaxation rate $\sigma$. This appears to be a manifestation of the proportionality between $T_c$ and the Fermi energy, which suggests a high energy scale for the SC coupling, and which is not consistent with the weak coupling of phonon-mediated SC. Studies of LaCuO and YBaCuO “parent” compounds show clear evidence of antiferromagnetism. YBa$_2$Cu$_{3-x}$CO$_x$O$_7$ shows the simultaneous presence of spin-glass magnetic ordering and superconductivity. The three-dimensional SC, (Ba,K)BiO$_3$, unlike the layered CuO-based compounds, shows no suggestion of magnetic ordering. Experimental techniques and theoretical implications will be discussed.
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The Muon Spin Rotation Technique

The use of the positive muon as a probe of condensed matter was first suggested by Garwin, Lederman and Weinrich in their 1957 paper describing their experimental observation of parity violation in muon production and decay. The positive muon is one of a number of light positive ions which have been used to probe condensed matter. These include the positron (e⁺), hole (h⁺), muon (μ⁺), pion (π⁺), proton (p⁺), deuteron (d⁺) and triton (t⁺). The muon is the least easily produced of all these particles, but the details of its behavior in condensed matter are far more easily discerned than for any other particles. This is because of subtleties in the nature of the weak interaction. Because of the parity violation inherent in the μ⁺ production,

\[ \pi^+ \rightarrow \mu^+ + \nu_\mu, \]

muons resulting from pion decay are 100% polarized in the center-of-mass frame. Consequently, by appropriate momentum selection (with magnets) one can obtain a nearly 100% polarized muon beam regardless (within reasonable limits) of the momentum of the source pions.

In like manner the muon decays into a positron through the weak interaction,

\[ \mu^+ \rightarrow e^+ + \nu_e + \bar{\nu}_\mu. \]

The positrons are emitted asymmetrically with respect to the muon spin, and with considerable kinetic energy (average: 35 MeV).

Data are taken with an apparatus such as the one our group uses at the Alternating Gradient Synchrotron of Brookhaven National Laboratory (Figure 1). Signals in scintillators 1, 2 and M5 with no signals in F6 indicate that a muon has stopped in the sample, which is in a thin-walled container (usually a cryostat). Signals in A4 and A3 (or in F7 and F8) indicate the emission of a positron in the backward (or forward) direction. The muon signal starts a clock, a positron signal stops it, and two histograms (backward and forward) are accumulated. A pair of magnet coils provides an external transverse field. The muons precess at a rate directly proportional to the magnetic field they sense, and this precession signal decays at a rate which depends upon the variance in the magnetic field observed by all the muons in the ensemble.

The data are fit to a function of the general form:

\[ N(t) = N_0 e^{-t/\tau} \left[ 1 + A G_x(t) \cos(\omega t + \phi) \right] + B, \]

where \( \tau = 2.2 \) microseconds (mean muon lifetime), \( A \) is the e⁺ anisotropy, \( G_x(t) \) is the depolarization function, \( \omega \) is the precession rate, \( \phi \) is a geometrical phase angle, and \( B \) is random background. \( A, G_x(t) \) and \( \omega \) are usually the quantities of physical interest.
MuSR Studies of HTSC Materials

The high-temperature copper-oxide-based superconductors discovered since the initial breakthrough on LaSrCuO by Bednorz and Müller, which have transition temperatures ranging from about 40K up to about 125K, are some form of type II superconductors. Type II's have a surface-to-volume energy relation which favors the intrusion of external applied magnetic fields in flux quanta which generally take on a uniform triangular geometry. This produces a variation in the internal magnetic field sensed by microscopic probes implanted into the material, such as $\mu^+$. This variation produces a depolarization of the muon spin precession, as in Figure 2, which shows the experimental asymmetry we obtained in YBa$_2$Cu$_3$O$_7$, both above and below its transition temperature. From the depolarization rate observed in the superconducting state, the magnetic penetration depth can be calculated. By combining the values from $\mu$SR with more conventional measurements, the superconducting charge carrier density and the effective mass of the carriers ($m^*/m_e$) may also be determined.

The variation in the magnetic field in a type II sample is given by

$$B(x) = B_0 \exp \left(-\frac{x}{\lambda_L}\right)$$

Our data show that, to a good approximation, the depolarization rate $\sigma$ is proportional to $T_c$. Relating this to the penetration depth gives

$$\sigma \propto \lambda_L^{-2} \propto \frac{n_s}{m^*} \propto T_c.$$

This suggests that the carrier density $n_s$ plays a major role in determining $T_c$. Furthermore, this relation appears to be inconsistent with standard Bardeen-Cooper-Schrieffer (BCS) theory, and suggests a high energy scale for the coupling mechanism. Such a high energy scale is found in models based on a large transfer integral between the oxygen and neighboring Cu atoms, and is also expected in the resonating-valence-bond picture.
Figure 2. Asymmetry in normal (top) and superconducting (bottom) YBa$_2$Cu$_3$O$_7$.

**Magnetic Ordering in HTSC Materials**

Muon spin rotation also serves as an exceptionally effective probe of magnetic ordering in materials at the microscopic level, and we have utilized this aspect of $\mu$SR to observe magnetic ordering in materials very closely related to HTSC materials. A prime example of this is our study of LaCuO$_{4-y}$, which is "parent" to the HTSC LaSrCuO. Figure 3 shows our data taken with zero external field on LaSrCuO (top) and La$_2$CuO$_{4-y}$ (Bottom). No oscillations are present in the top case, but they are clearly observed in the lower plot. This indicates the presence of antiferromagnetism with a moment per Cu atom of about 0.5 Bohr magneton. A similar experiment done at TRIUMF on oxygen-depleted YBa$_2$Cu$_3$O$_{7-y}$ shows similar antiferromagnetic ordering in that material. These results suggest a close relationship between superconductivity and magnetic ordering in the new CuO-based HTSC materials, which is opposite to all experiences with conventional superconductors (heavy-fermion materials which have superconducting phases show close relations between SC and magnetism in some cases also).
Figure 3. Zero-field asymmetry in LaSrCuO and LaCuO_{4-y}.
AFM is observed in the lower spectrum.

A study of YBaCuO doped with Co, YBa_{2}Cu_{3-y}Co_{x}O_{7}, shows simultaneous SC and magnetic ordering, in this case the spin-glass variety in which magnetic moments are frozen into random but static orientations. For x = 0.1, for example, the depolarization rate begins to rise at about 40K, indicating the onset of SC, while the initial polarization begins to decrease at about 30K, indicating the onset of spin-glass ordering. Figure 4 shows how the SC transition temperature T_{c} and the magnetic ordering transition temperature T_{m} vary with Co concentration. There is a clear overlap of SC and spin glass ordering. We suspect that this does not violate the Meissner effect because the two effects are probably occurring in microscopically distinct regions of the sample, such as alternating planes.

In light of the magnetic ordering present in materials closely related to the essentially two-dimensional CuO perovskite HTSC's, we studied three-dimensional perovskite superconductors which do not contain Cu, (Ba,K) BiO_{3}. These showed no evidence for magnetic ordering, which suggests that these materials are probably not likely candidates for record-setting values of T_{c}.
Transition temperatures as functions of Co concentration in YBa$_2$Cu$_{3-x}$Co$_x$O$_y$.

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