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Y. J. Uemura (1), B. J. Sternlieb (1), D. E. Cox (2), V. J. Emery (2), A. Moodenbaugh (2), M. Suenaga (2), J. H. Brewer (3), J. F. Carolan (3), W. Hardy (3), R. Kadono (3), J. R. Kempton (3), R. F. Kieff (3), S. R. Kreitzman (3), G. M. Luke (3), P. Mulhern (3), T. Riseman (3), D. L. Williams (3), B. X. Yang (3), W. J. Kossler (4), X. H. Yu (4), H. Schone (4), C. E. Stronach (5), J. Gopalakrishnan (6), M. A. Subramanian (6), A. W. Sleight (6), H. Hart (7), K. W. Lay (7), H. Takagi (8), S. Uchida (8), Y. Hidaka (9), T. Murakami (9), S. Etemad (10), P. Barboux (10), D. Keane (11), V. Lee (12), and D. C. Johnston (13)

(1) *Department of Physics, Columbia University, New York, NY 10027, U.S.A.*
(2) *Brookhaven National Laboratory Upton, New York 11973, U.S.A.*
(3) *TRIUMF and Univ. of British Columbia, Vancouver, B.C., V6T 2A3, Canada*
(4) *College of William and Mary, Williamsburg, Virginia 23185, U.S.A.*
(5) *Virginia State University, Petersburg, Virginia 23803, U.S.A.*
(6) *Du Pont Experimental Station, Wilmington, Delaware 19898, U.S.A.*
(7) *G.E. Corporate Research and Development, Schenectady, New York 12301, U.S.A.*
(8) *University of Tokyo, Hongo, Tokyo 113, Japan*
(9) *NTT Optoelectronics laboratory, Tokai, Ibaraki 319-11, Japan*
(10) *Bell Communications Research, Red Bank, New jersey 07701, U.S.A.*
(11) *IBM T. J. Watson research Center, Yorktown Heights, New York 10598, U.S.A.*
(12) *IBM Almaden Research Center, San Jose, California 95120, U.S.A.*
(13) *Ames Laboratory (USDOE), Ames, Iowa 50011, U.S.A.*

Abstract. – Recent progress of our μSR measurements on high-T_C and related systems is presented. Static magnetic order has been found and studied in La_2CuO_4, $(La_{2-x}Sr_x)CuO_4$, $YBa_2Cu_3O_x$, $YBa_2(Cu_{3-x}Co_x)O_7$, as well as in $Bi_2Sr_2YCu_2O_8$. No magnetic order was found in the 3-dimensional systems $BaBiO_3$, $Ba(Pb_{0.1}Bi_{0.9})O_3$, and (BaK) BiO_3 above $T = 10$ K. The anisotropy of the magnetic field penetration depht λ has been observed by using a c-axis alligned sintered pellet specimen of $YBa_2Cu_3O_7$. The temperature dependence of $\lambda_{\parallel}$ (for $c \parallel H_{ext}$) indicates isotropic superconducting energy gap without any anomalies. Based on the muon spin relaxation rate σ observed in (random) pellet specimens of various high-T_C superconductors, we found a linear relation between T_C and $\sigma \propto 1/\lambda^2 \propto n_s/m^*$, which suggests a high energy scale of superconducting coupling. Deviations from this linear relation are seen for samples with large carrier concentrations n_s.

1. Introduction

The discovery of high-T_C suprconductors [1] has brought a new excitement to scientists working in the field of magnetism, because the typical oxide high-T_C systems, $(La_{2-x}Sr_x)CuO_4$ and $YBa_2Cu_3O_x$, are associated with family compounds which exhibit magnetic order. For the study of the superconducting and magnetic properties of high-T_C systems, muon spin relaxation (μSR) has been used extensively by various groups [2-12], and found to be very informative. In this paper, we describe the most recent progress of our ongoing μSR studies at the TRIUMF and AGS/BNL muon channels in order to illustrate how μSR probes the interplay between magnetism and superconductivity in these systems.

2. Experimental methods

A schematic view of the basic principles of the μSR technique is shown in figure 1 of reference [13]. A spin polarized positive muon beam is stopped in a specimen, and time histograms of muon-decay positrons are collected. Since positrons are emitted preferentially in the muon spin direction, the time evolution of the muon spin polarization can be measured directly *via* the angular asymmetry in the positron counting rate.

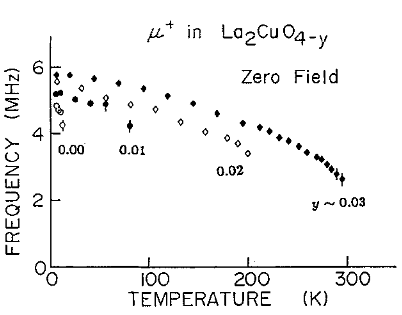

Fig. 1. – Muon spin precession frequency ν_μ observed in zero field on various sintered powder specimens of La_2CuO_{4-y}. The relative precision of y is $\Delta y \leq 0.005$, while the absolute values of the stoichiometry of La, Cu, and O are subject to larger systematic errors (from Uemura *et al.* [4]).

This measurement can be performed even without applying an external magnetic field (zero-field μSR; ZF-μSR).

When a transverse external magnetic field $H_{\rm ext}$ is applied perpendicularly to the initial polarization direction of the muons, the muon spins precess around $H_{\rm ext}$ and a sinusoidal signal appears in the positron time spectra. This transverse-field μSR (TF-μSR) method is used in the study of the magnetic-field penetration depth in type-II superconductors.

In the ordered state of uniform ferro- or antiferromagnetic materials, a finite static internal field $H_{\rm int}$ is generated at the muon site by the neighbouring ordered moments. In polycrystalline specimens, the direction of $H_{\rm int}$ is random but the magnitude $|H_{\rm int}|$ is often unique. Then one sees the sinusoidal oscillation signal of the muon spins precessing around $H_{\rm int}$ in the absence of an applied field. If the spatial spin structure is random, as in spin glasses, $H_{\rm int}$ has a wide distribution, and the oscillation around $H_{\rm int}$ is rapidly damped [14]. One observes a characteristic depolarization function in ZF-μSR in this case.

μSR has a superb sensitivity to static magnetic order. Even an ordered moment as small as 0.01 μ_B per formula unit in either a uniform or random spin configuration can be detected. μSR is also a volume sensitive method. Each of the magnetically ordered, paramagnetic, or superconducting states produces a different type of signal. Therefore, one can often estimate the volume fraction of the different phases in a specimen accurately. These features are helpful in obtained magnetic phase diagrams. (For general aspects of μSR, see Ref. [15].)

3. Magnetic order and phase diagrams

1) La_2CuO_{4-y}. – La_2CuO_4 is the antiferromagnetic parent compound of the high-T_C systems (LaSr) CuO_4 and (LaBa) CuO_4. In early 1987, we found [2] a long lived muon spin precession signal in zero field in La_2CuO_{4-y} with $y = 0.02 \sim 0.03$ below the Néel temperature. An example of the precession signal is shown in figure 1 of reference [2]. The frequency ν_μ of this precession is proportional to the static internal magnetic field $H_{\rm int}$, and consequently to the magnitude of the ordered moments on the Cu atoms around the muon site. Figure 1 of the present paper shows the temperature dependence of ν_μ observed [4] in four different specimens of La_2CuO_{4-y}.

With increasing oxygen concentration, the ordering temperature T_N is sharply reduced. The frequency $\nu_\mu (T \to 0)$, however, is not much changed. μ^+ is a point-like local probe, and the internal field at the muon site is determined predominantly by a small number of magnetic moments near the muon site. Therefore, figure 1 indicates that the magnitude of the local ordered moment S on each Cu atom in the ground state is almost independent of the oxygen concentration.

When we combine this μSR result with neutron scattering studies [16], which are sensitive to longer-range spatial spin correlations (see Ref. [4]), we find that the spin correlations become short ranged upon oxygen doping, i.e., upon introduction of holes into the system. We can also estimate the magnitude of ordered moment to be $S \sim 0.5\ \mu_B$ based on the combined neutron and muon studies. This value can be explained either in terms of a 2-dimensional spin 1/2 Heisenberg model for localized moments or by spin density waves for more itinerant electrons.

2) $(La_{2-x}Sr_x)\,CuO_4$. – The substitution of Sr^{2+} for La^{3+} introduces holes into the system, as does doping with O^{2-}. In the $x = 0.025$ sample of $(La_{2-x}Sr_x)\,CuO_4$, we found the onset of static magnetic order occurs around $T = 10$ K; a clear muon spin precession signal was observed below $T \sim 5$ K. In the μSR measurements, this sample looks more like a "short-range antiferromagnet" than a spin glass. It is not clear if there is a distinct phase boundary between antiferromagnetic and spin glass phases or if the increasing randomness in the spatial spin correlations with increasing x gradually leads from antiferromagnet to spin glass states.

We also found that a $x = 0.07$ sample, which is superconducting below $T_C \sim 16$ K, starts static magnetic order around $T \sim 5$ K. In single crystal specimens, low superconducting transition temperatures $T_C \sim 10$ K suggest that the real hole concentration may be smaller than the nominal concentration. In a superconducting single crystal ($T_C \sim 10$ K, real hole concentration is presumably equivalent to $x \sim 0.06$) we found, using μSR, that almost the entire volume undergoes static magnetic order (or freezing) below $T \sim 4$ K.

3) $YBa_2Cu_3O_x$. – μSR was also applied to the family compounds $YBa_2Cu_3O_x$ of the 90 K superconductor $YBa_2Cu_3O_7$. Nishida *et al.* [7] first found static magnetic order in $x = 6.2$ system by μSR. Subsequently, the antiferromagnetic spin structure was established by neutron scattering studies [17]. We have performed [3] detailed μSR measurements on this series at TRIUMF. Figure 2 shows the phase diagram we determined on the specimens prepared by rapid quenching from high temperatures. The ordering temperature decreases sharply with increasing oxygen concentration x, and the 3-dimensional static magnetic order disappears around $x = 6.4 \sim 6.5$ where superconductivity sets in. The muon spin precession frequency ν_μ is almost constant between $x = 6.0$ and 6.35, indicating that the ordered moment of Cu is independent of x. Around $x \sim 6.4$, we observed ZF-μSR spectra resembling those found in the spin glasses [14] CuMn and AuFe. This reflects the increasingly random distribution of internal fields with increasing x.

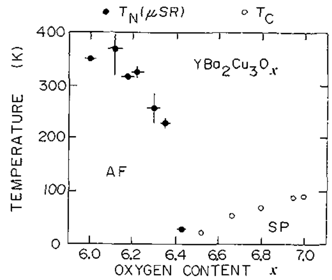

Fig. 2. – Phase diagram of $YBa_2Cu_3O_x$ (specimens produced by rapid quenching) plotted against the oxygen concentration x. The closed circles stand for the Néel temperatures T_N determined by our μSR measurements (from Brewer *et al.* [3]), while open circles represent superconducting transition temperatures T_C determined by various methods. It is shown that the superconducting phase (SP) sets in around the oxygen concentration where the 3 dimensional magnetic order in the antiferromagnetic phase (AF) disappears.

4) $YBa_2(Cu_{3-x}Co_x)O_7$. – In the $YBa_2(Cu_{3-x}Co_x)O_7$ system, the superconducting transition temperature T_C decreases with increasing Co concentration x as shown in figure 3. Using μSR, we have recently found [18] that almost the entire volume of the specimen undergoes static magnetic order below $T \sim 15$ K for the $x = 0.1$ system ($T_C \sim 60$ K), and below $T \sim 20$ K for the $x = 0.2$ system ($T_C \sim 40$ K). There is no signature of the magnetic transition in susceptibility of the $x = 0.1$ and 0.2 specimens which shows the diamagnetic susceptibility below T_C. At the moment, it is not certain whether only the Co moments or both Co and Cu moments participate in the magnetic order. One possible scenario is that Co occupies the so-called chain Cu sites and mediates a strong magnetic coupling between the two adjacent CuO planes, playing the role of a catalyst in the ordering of the Cu moments.

5) $Bi_2Sr_2YCu_2O_8$. – By substituting Y^{3+} ions for Ca^{2+} ions in the high-T_C superconductor $Bi_2Sr_2CaCu_2O_8$, one can reduce the number of carriers and make an insulating specimen. Quite recently, we have performed ZF- and TF-μSR measurements on this $Bi_2Sr_2YCu_2O_8$ specimen, and found that the material undergoes the static magnetic order with a broad distribution of transition temperature around $T \sim 200$ K. Figure 4 shows the muon decay asymmetry observed in zero field at $T = 3.7$ K. We see a clear signal of muon spin precession around static internal fields H_{int} (at least two different frequencies are seen). This result established that the family compound of BiSrCaCuO orders magnetically, just like the family systems of (LaSr)CuO_4 and $YBa_2Cu_3O_7$.

6) (BaK)BiO_3 and Ba(PbBi)O_3. – Superconductivity with $T_C \sim 30$ K has recently been found [19] in the 3-dimensional (3-d) perovskite system (BaK)BiO_3. This system has a 3-d BiO network instead of the 2-dimensional (2-d) CuO planes found in other high-T_C systems with layered crystal structures. There are, however, some key features shared by the 2-d and 3-d systems; e.g., the parent compounds La_2CuO_4 and $BaBiO_3$ are both half-filled band systems on average ($Ba^{2+}Bi^{4+}O_3^{6-}$ leaves one of the five valence electrons of Bi on each Bi atom), and the doping of K or Sr introduces a charge carrier (a hole) into the system. Using μSR, we have searched for static magnetic order in the family systems of (BaK)BiO_3.

No signature of magnetic order was found in $BaBiO_3$ (monoclinic crystal structure) above $T = 10$ K, in $Ba(Pb_{0.1}Bi_{0.9})O_3$ (orthorhombic) above $T = 10$ K, or in non-superconducting (BaK)BiO_3 (nearly cubic) above $T = 3.4$ K. In a superconducting specimen

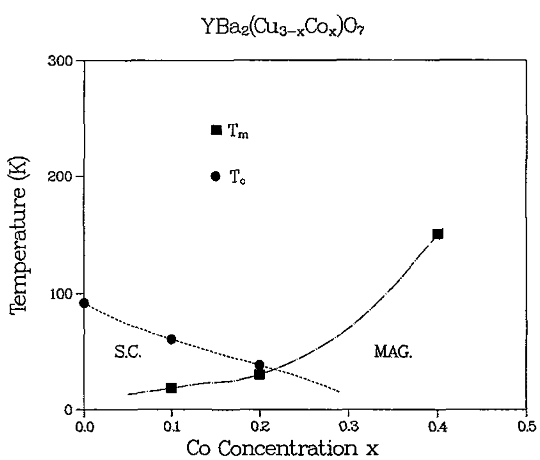

Fig. 3. – Phase diagram of the $YBa_2(Cu_{3-x}Co_x)O_7$ system *determined by the μSR measurements (from* Yu *et al.* [18]). S.C. denotes the superconducting state, and MAG denotes the state with static magnetic order.

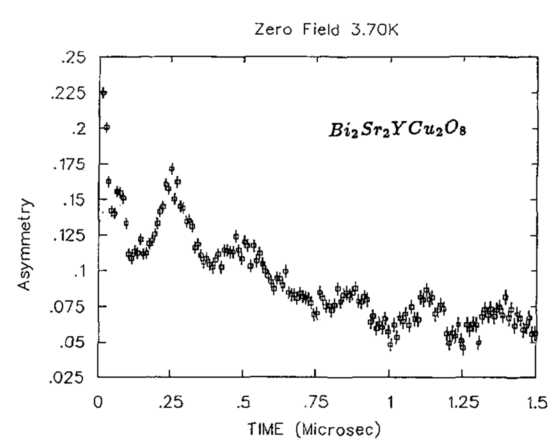

Fig. 4. – Asymmetry of muon decay signal observed in μSR measurement on a sintered pellet specimen of $Bi_2Sr_2YCu_2O_8$ at $T = 3.7$ K *in zero field*. The observed muon spin precession demonstrates that this material undergoes static magnetic ordering.

of (BaK) BiO_3 ($T_C = 29$ K), we found by μSR that about 20 % of the total volume is superconducting, while the remaining 80 % of the volume is either non-magnetic or paramagnetic above $T = 4.6$ K. The absence of the static magnetic order in the family compounds of (BaK)BiO_3 is in a sharp contrast to the 2-d systms where superconductivity appears in the vicinity of magnetic order. This may be due to the more itinerant character of the Bi 6s electrons compared to the Cu 3d electrons.

4. Magnetic-field penetration depth

To measure the magnetic field penetration depth λ in type-II superconductors, we apply a transverse external magnetic field H_{ext} ($H_{C1} \leq H_{ext} \leq H_{C2}$) in a direction perpendicular to the initial muon spin polarization. Typical muon spin precession signals above and below T_C are shown in figure 4 of reference [13]. Above T_C, the muon spins are not depolarized because H_{ext} penetrates the sample uniformly. Below T_C, H_{ext} penetrates by forming a lattice of flux vortices. The local field at a muon site is largest when the site is close to a flux core, and smallest between the vortices. Hence there is a width ΔH in the local field which causes muon spin depolarization. Since ΔH is determined by the penetration depth λ as $\Delta H \propto 1/\lambda^2$, we can directly deduce the values of λ from the muon spin relaxation rate σ which is proportional to ΔH. For simplicity, we fitted our data by using a Gaussian depolarization function $\exp\left(-0.5\,\sigma^2 t^2\right)$.

We have performed μSR measurements on a sintered pellet specimen of $YBa_2Cu_3O_7$ which was fabricated with the orientation of the c-axes of the microcrystallates alligned within 9° HWHM. Figure 5 shows the muon spin relaxation rate σ observed in $H_{ext} = 13$ kG for three different orientations of c-axis with respect to H_{ext}. There is clear anisotropy due to the difference between $\lambda_{\parallel}$ (for $c \parallel H_{ext}$) and $\lambda_{\perp}$ (for $c \perp H_{ext}$).

When the c-axis is parallel to H_{ext}, the current circulating within the CuO planes screens H_{ext} most effectively, and the penetration depth is the shortest. In this configuration, the detailed distribution of the local field $P(H)$ was deduced from the observed line shape of the muon depolarization function. $P(H)$ has an asymmetric shape with a larger spectral weight in the high frequency side, being consistent with the field distribution expected for a triangular Abrikosov lattice of flux vortices.

Based on an approximate relation [20] between ΔH and λ (modified for triangular lattice; see Ref. [6]), we have deduced the hard-axis penetration depth $\lambda_{\parallel}$ from the relaxation rate σ for $c \parallel H_{ext}$.The temperature dependence of $\lambda_{\parallel}$ is shown in figure 6. The absolute value $\lambda_{\parallel}(T \to 0)$ is about 1460 Å. The temperature dependence agrees well with the so-called empirical formula

$$\lambda(T) = \lambda(T=0)\,/\sqrt{1-(T/T_C)^4}$$

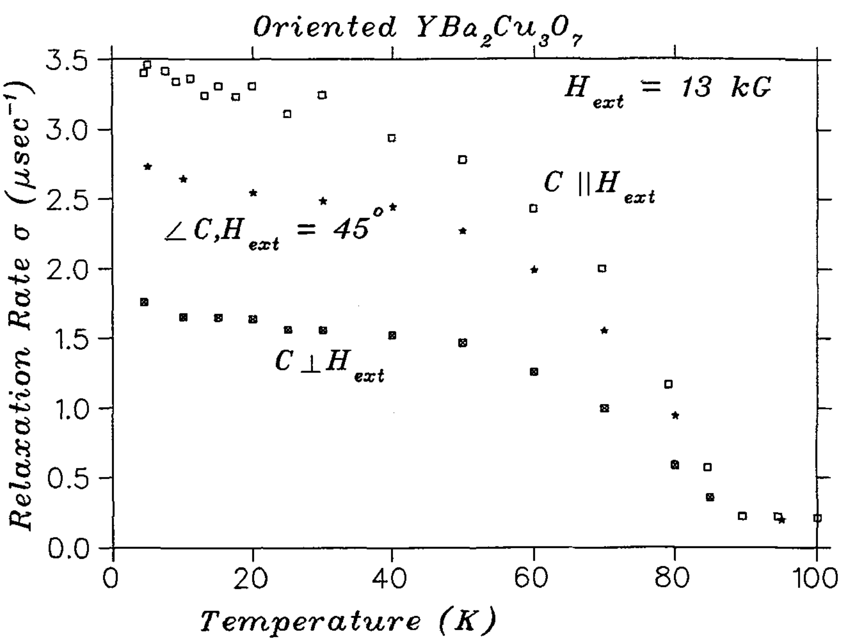

Fig. 5. – Muon spin relaxation rate σ observed in the TF-μSR measurements with H_{ext}=13 kG in the c-axis aligned pellet specimen of $YBa_2Cu_3O_7$. The relaxation rate is largest when we applied H_{ext} parallel to the c-axis. This confirms the anisotropy of the penetration depth λ.

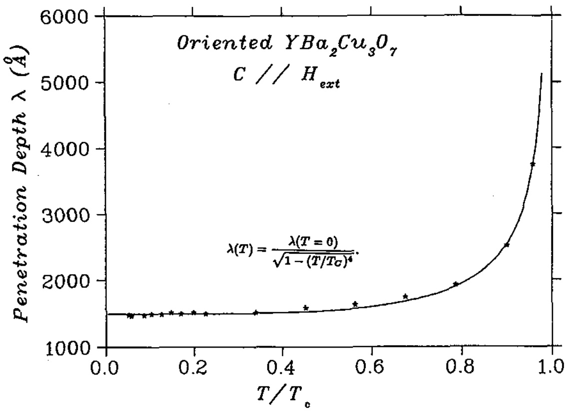

Fig. 6. – Temperature dependence of the magnetic field penetration depth $\lambda_{\parallel}$ derived from the muon spin relaxation rate σ in a c-axis oriented sintered specimen of $YBa_2Cu_3O_7$ with the external field H_{ext} applied parallel to the c-axis. The solid lines represent fits of the data to the so-called empirical formula $\lambda(T) = \lambda(T=0)/\sqrt{(1-(T/T_C))^4}$.

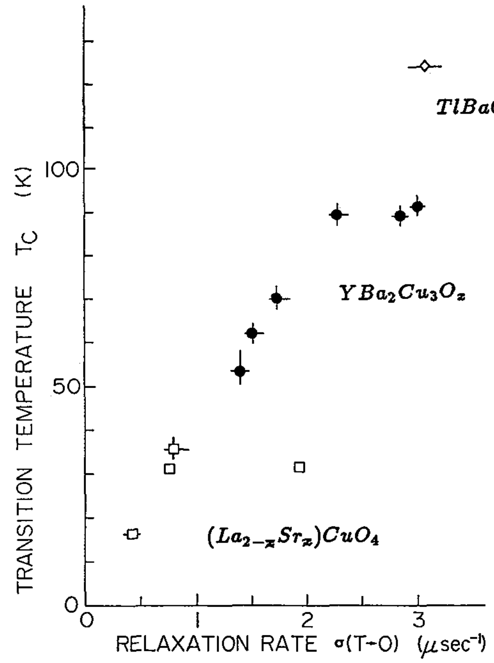

Fig. 7. – Superconducting transition temperatures T_C, as determined by μSR measurements, plotted against the values of the muon spin relaxation rate $\sigma(T \to 0)$ for non-aligned pellet specimens of high-T_C superconductors $(La_{2-x}Sr_x)CuO_4$, $YBa_2Cu_3O_x$, and TlBaCaCuO. A linear relation between T_C and $\sigma(T \to 0) \propto 1/\lambda^2 \propto n_s/m^*$ is seen for the initial increase of $\sigma(T \to 0)$, while deviations from the linear line is noticed to take place at system-dependent values of σ.

which is expected for superconductors with an isotropic superconducting energy gap. This formula exhibits a slower change around $T \to 0$ with increasing temperature than the solution of the isotropic BCS model with a short coherence length [21]. Therefore, we can safely say that there is no anomaly in the energy gap, and that the coupling of superconducting carriers has s wave symmetry.

In μSR measurements on randomly oriented powder or pellet specimens, some sort of directionally averaged values of λ are obtained. We can, however, compare the results on various different high-T_C systems on the assumption that the process of directional averaging is not different for different systems. The muon spin relaxation rate σ is proportional to $1/\lambda^2$, and consequently to the superconducting carrier concentration n_s divided by the effective mass m^* as

$$\sigma \propto 1/\lambda^2 \propto n_s/m^*.$$

Therefore, by plotting the values of T_C against the observed results of $\sigma(T \to 0)$, we can study the relation between the transition temperature and n_s/m^*.

We have accumulated μSR data on superconducting (random) pellet specimens of $(La_{2-x}Sr_x)CuO_4$ (x = 0.07, 0.1, 0.15, 0.2), $YBa_2Cu_3O_x$ (x = 6.6 ~ 7.0), and $Tl_2Ba_2Ca_2Cu_3O_x$ (T_C=125 K) systems. Figure 7 shows the plot of T_C *versus* $\sigma(T \to 0)$ obtained in these systems. From the initial rise of T_C with increasing $\sigma \propto n_s/m^*$, we found that a linear relation holds between T_C and $\sigma(T \to 0)$ regardless of sample composition. As discussed in references [6, 13], this linear relation can be understood as the manifestation of the relation $T_C \propto \varepsilon_F$ (ε_F is the Fermi energy) which is expected when the superconducting coupling has a high energy scale. The linear relation $T_C \propto n_s/m^*$ can not be expected in the so called weak coupling limit of the phonon-mediated BCS superconductors.

We also found that each series of compounds exhibits a deviation from this linear relation when $\sigma(T \to 0)$ exceeds some system-dependent characteristic value. This feature is most remarkably demonstrated in $(La_{1.9}Sr_{0.1})CuO_4$ (T_C~30 K) and $(La_{1.8}Sr_{0.2})CuO_4$ (T_C~30 K) where we obtained a value of $\sigma(T \to 0)$ for the $x = 0.2$ specimen twice that found for the $x = 0.1$ specimen. The superconducting transition temperature of the $(La_{2-x}Sr_x)CuO_4$ system is highest when $x = 0.15$, and is known to decrease with increasing x in the region $x \geq 0.15$. The μSR results indicate that the number of superconducting carriers is still increasing linearly to x when $x \geq 0.15$, so T_C is suppressed for some other reasons. This tendency of "T_C saturation" is also seen in the results on the $YBa_2Cu_3O_x$ series in figure 7.

5. Discussions

In the antiferromagnetic family of the high-T_C systems, the reduction of T_N with increasing O or Sr concentration can be understood as the result of an increasing number of frustrated bonds in the CuO planes (see Ref. [13]). The problem of the coexistence of superconductivity and magnetic order in the CuO planes still remains open to further detailed study, because the difficulties related to the spatial inhomogeneity of the dopant concentration have not yet been fully overcome. We have presented a few candidate cases which suggest that the static magnetic order of Cu moments and superconductivity may coexist.

Very recently, we have discovered static magnetic order in a family compound of BiSrCaCuO system in which Ca atoms are substituted by Y atoms to reduce the carrier concentration. Therefore, it seems that superconductivity occurs in the vicinity of magnetic order in all the 2-dimensional layered high-T_C systems characterized by CuO planes. This suggests a deep relation between magnetism and superconductivity. In contrast, the absence of magnetic order in the 3-d (BaK) BiO_3 and family systems indicates that static magnetic order in family systems is not a necessary condition to obtain a high transition temperature.

In the study of superconductors, we learned that the superconducting energy gap is finite without any anomalies, and that the energy scale of superconducting coupling may be very high. It is a challenging problem to explain the observed saturation and reduction of T_C with increasing carrier concentration n_s/m^*. Ongoing μSR studies will provide additional information for the better understanding of high-T_C superconductivity.

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