Basic Similarities among Cuprate, Bismuthate, Organic, Chevrel-Phase, and Heavy-Fermion Superconductors Shown by Penetration-Depth Measurements

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Muon-spin-relaxation and bulk measurements of the magnetic-field penetration depth suggest that the cuprate high-T_c, bismuthate, organic, Chevrel-phase, and heavy-fermion systems possibly belong to a unique group of superconductors characterized by high transition temperatures T_c relative to the values of n_s/m^* (carrier density/effective mass). This feature distinguishes these exotic superconductors from ordinary BCS superconductors.

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The magnetic-field penetration depth λ is one of the most fundamental parameters of superconductivity. In superconductors close to the clean limit, $1/\lambda^2$ is determined essentially by the superconducting carrier density n_s divided by the effective mass m^*. Thus, one can compare n_s/m^* of different superconductors by measuring λ. The muon-spin-relaxation (μSR) technique provides a direct method to measure λ in type-II superconductors.[1-3] Absolute values of λ can often be determined with much better accuracy by μSR than by other bulk techniques, such as magnetization or H_{c1} measurements which require detailed knowledge about the surface area or flux pinning. Taking advantage of this feature, we demonstrated by μSR in a previous Letter[4] that remarkable correlations exist between T_c and n_s/m^* in many different cuprate high-T_c superconductors.

In this paper, with our new μSR results on bismuthate $(Ba_{1-x}K_x)BiO_4$ (BKBO), Chevrel-phase, and organic $(BEDT\text{-}TTF)_2Cu(NCS)_2$ (BEDT) superconductors, we show that these systems also follow the same correlations between T_c and n_s/m^* found for the cuprate systems. We suggest that even some heavy-fermion (HF) superconductors may follow these correlations, and demonstrate a clear contrast between these systems and ordinary type-II superconductors like Nb. By combining the μSR results with the Sommerfeld constant γ, we further consider relations among T_c, the Fermi energy ϵ_F, the thermal wavelength $\Lambda(T)$, and the Bose-Einstein condensation temperature T_B, and illustrate implications of these correlations in a phenomenological approach.

In μ^+SR measurements of the penetration depth, a beam of spin-polarized positive muons is stopped in a specimen (typically 2 cm in diameter, 2 mm thick), and the muon-decay positrons are detected individually to accumulate the time histogram $N(t)$ of more than a million decay events. Usually, an external magnetic field H_{ext} is applied along the beam direction $\hat{z}$, with the initial muon spin direction perpendicular to $\hat{z}$. Since positrons are emitted preferentially along the muon spin direction, the muon spin precession around H_{ext} produces a sinusoidal oscillation in the time histogram:

$$N(t)=N_0\exp(-t/\tau_\mu)[1+AG_x(t)\cos(\omega t+\phi)]\,,$$

where $\tau_\mu=2.2$ μs is the muon lifetime, $A\sim0.2$ is the initial asymmetry, and the relaxation function $G_x(t)$ describes the depolarization of muon spins.[1]

In the superconducting state of type-II superconductors with $H_{c1}\ll H_{\text{ext}}<H_{c2}$, the external field penetrates into the specimen by forming a lattice of flux vortices. This results in an inhomogeneous width ΔB of the local fields which causes the depolarization. Usually $G_x(t)$ is approximated by a Gaussian form $\exp(-\sigma^2t^2/2)$ with the relaxation rate σ proportional to ΔB. It is known that ΔB is proportional to $1/\lambda^2$ and nearly independent of H_{ext} over a wide range of the field.[5] Generally, $1/\lambda^2$ is a function of n_s, m^*, the coherence length ξ, and the mean free path l, leading to

$$\sigma\propto\Delta B\propto\frac{1}{\lambda^2}=\frac{4\pi n_s e^2}{m^*c^2}\frac{1}{1+\xi/l}\,.$$

Based on currently available estimates[6] of ξ and l from H_{c2}, resistivity, and de Haas-Shubnikov oscillation studies, one finds that typical cuprate ($YBa_2Cu_3O_y$ with $y=6.7\text{-}7.0$), organic BEDT, Chevrel-phase, and HF (UPt_3) systems satisfy $\xi/l\leq0.3$. Although no accurate estimate is available for l in BKBO and some other systems, it is reasonable to assume little effect from ξ/l in most of the systems described in this paper in view of their short coherence lengths ξ. Therefore, we can use the muon-spin-relaxation rate σ as a measure of n_s/m^*.

The μSR measurements on $(Ba_{1-x}K_x)BiO_3$, with $x=0.4,0.5$, and Chevrel-phase $LaMo_6Se_8$ ($T_c=11$ K), $LaMo_6S_8$ ($T_c=7$ K), and $PbMo_6S_8$ ($T_c=14$ K) systems were carried out at TRIUMF (Vancouver) using polycrystalline specimens prepared as described elsewhere.[7] Figure 1(a) shows the temperature dependence of the relaxation rate σ observed with the transverse external field $H_{ext}\sim 2$ kG in the field-cooling measurements. In both BKBO and Chevrel-phase systems, $\sigma(T)$ shows a sharp increase below T_c with saturation in the low-temperature region, and agrees well with the formula of the two-fluid model (solid lines). These features are characteristic of s-wave pairing without nodes in the energy gap, and are common to cuprate high-T_c superconductors.

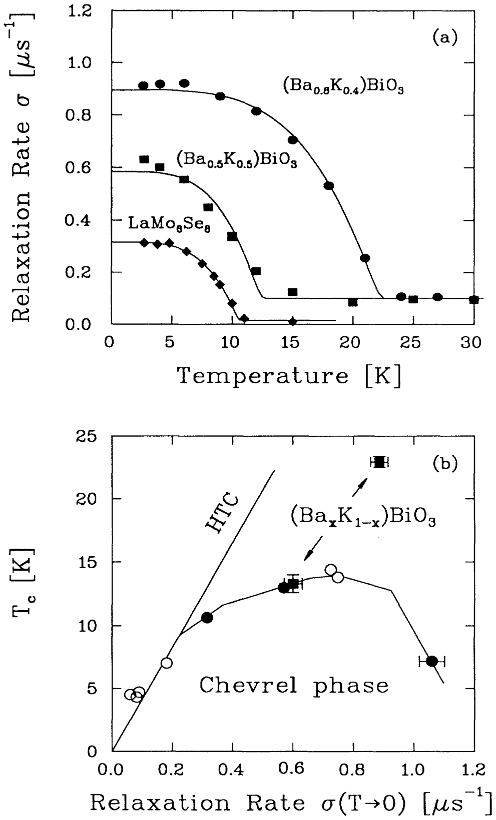

FIG. 1. (a) Temperature dependence of the muon-spin-relaxation rate σ observed in polycrystalline specimens of $(Ba,K)BiO_3$ and a Chevrel-phase superconductor $LaMo_6Se_8$. The field-cooled data agree well with the two-fluid model shown by the solid lines. (b) A plot of T_c vs $\sigma(T\rightarrow 0)$ of the BKBO (solid squares) and Chevrel systems [solid circles from the present study and open circles from Birrer *et al.* (Ref. 8)]. The straight line corresponds to the linear relation found for the cuprates.

In Fig. 1(b), we plot the low-temperature relaxation rate $\sigma(T\rightarrow 0)$ vs T_c as determined by μSR in the BKBO and Chevrel systems. In addition to the present results, we also included the earlier μSR results by Birrer *et al.*[8] on some other Chevrel-phase compounds $SnMo_6S_ySe_{8-y}$ and $PbMo_6S_ySe_{8-y}$ (open circles). Note that all the Chevrel systems described here are nonmagnetic ones. In Chevrel systems, T_c initially increases with increasing σ, then shows saturation and suppression. This feature is remarkably similar to the tendency found in cuprate high-T_c systems. Moreover, the initial increase of T_c follows the linear relation with the slope identical to that found in the cuprate systems. The points from the BKBO systems also lie relatively close to this line.

In Fig. 2, we compare the results in Fig. 1(b) with those obtained in ceramic specimens of the high-T_c cuprate superconductors.[4,9] In highly two-dimensional (2D) cuprate systems with anisotropic penetration depth, the polycrystalline results for σ predominantly reflect[10] the in-plane penetration depth λ_{in} which is defined for $H_{ext}\perp CuO_2$ planes. We have also included our new results[3,11] on the organic BEDT system obtained with H_{ext} applied perpendicular to the conductive b-c plane, after making a $\sim 40\%$ correction[10] to account for the difference between the single-crystal and polycrystalline results. We see that Chevrel, BKBO, and BEDT systems give points lying close to those of cuprates. This figure suggests a possibility that all of these systems belong to a special group of superconductors with some fundamental features in common. This aspect becomes clearer when

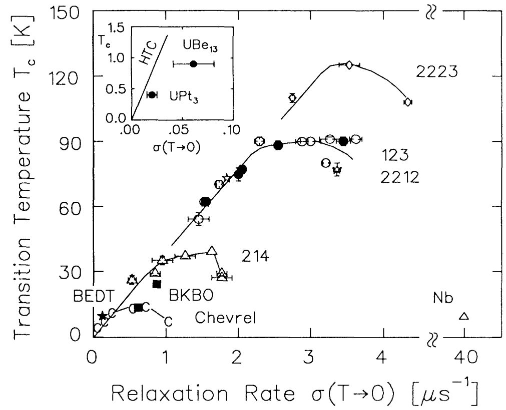

FIG. 2. Plot of T_c vs $\sigma(T\rightarrow 0)\propto n_s/m^*$ of cuprates (Refs. 4 and 9), BKBO (solid squares, present work), Chevrel-phase (C, present work and Ref. 8), and BEDT (solid star, present work) systems, based on the μSR measurements; and of Nb (Ref. 12) and HF systems (inset) (Ref. 16) with the values of σ estimated from the bulk penetration-depth measurements. The straight line in the inset corresponds to the linear relation found for the cuprates.

we include a point for Nb (deduced for magnetization results[12] of λ) which lies very far from other points in Fig. 2. In Nb and other ordinary type-II superconductors, T_c is low, whereas n_s/m^* is large mainly due to the large carrier density. In contrast, the cuprate, BKBO, BEDT, and Chevrel systems have relatively high T_c in spite of low n_s/m^*, with the former three systems characterized by low carrier density and the Chevrel systems by relatively large m^* in addition to low n_s.

A comparison with results on heavy-fermion systems should be interesting.[13] We have recently performed μSR measurements[3,14] in UPt_3 and UBe_{13}. In both of these systems, $\sigma(T)$ measured in $H_{\text{ext}}=2$–4 kG does not show any significant change below $T\sim 1.2T_c$. This indicates that the penetration depths in these systems are longer than 10000 Å, unfortunately beyond the sensitivity of the μSR technique. Although $\sigma(T)$ in UPt_3 measured in lower fields ($\sim$200 G) shows an increase below T_c (as reported also by Broholm *et al.*[15]), we consider the high-field results to be more reliable since they are much less sensitive to extrinsic effects. Our estimate $\lambda \geq 10000$ Å agrees well with the bulk flux confinement study of Gross *et al.*,[16] who reported $\lambda = 11000$ Å for UBe_{13} and 19000 Å for UPt_3. We converted these values into the expected μSR relaxation rates σ, and show them in the σ vs T_c plot (inset of Fig. 2). The points from these HF systems also lie close to the linear relation of the cuprate systems. This indicates that the HF systems have relatively high T_c as scaled with their $\sigma \propto n_s/m^*$, and thus possibly join the group of the cuprate and other superconductors. In the HF systems, it is the heavy effective mass m^* which results in a large λ and small σ.

These results for σ enable simple estimates of the Fermi energy ϵ_F. In the highly 2D cuprate and organic systems, we can deduce the 2D carrier density $n_{s\,2D}$ on the conductive planes from the interplane separation c_{int} and σ which reflects the 3D density n_s. Thus, we obtain $\epsilon_F=(\hbar^2\pi)n_{s\,2D}/m^*$ using the formula of noninteracting 2D electron gas. For 3D BKBO, Chevrel, and HF systems, we combine $\sigma \propto n_s/m^*$ with the observed values of the Sommerfeld constant $\gamma \propto n_s^{1/3}m^*$ to calculate

$$\epsilon_F=(\hbar^2/2)(3\pi^2)^{2/3}n_s^{2/3}/m^* \propto \sigma^{3/4}\gamma^{-1/4}$$

(see Ref. 17). Figure 3 shows a log-log plot of T_c vs ϵ_F/k_B thus estimated from the results of σ shown in Fig. 2. Corresponding values of $n_{s\,2D}/m^*$ in 2D and $n_s^{2/3}/m^*$ in 3D systems are indicated on the horizontal axis. Points from the cuprate, BEDT, BKBO, Chevrel, and HF superconductors follow a general linear trend common to all of these systems, which we shall henceforth call "exotic" superconductors. The T_c's of these exotic systems range between 1/100 and 1/10 of ϵ_F/k_B, in clear contrast to Nb, Sn, and other ordinary BCS superconductors (e.g., Al, Zn) which have T_c of less than 1/1000 of ϵ_F/k_B (see Fig. 3). This argument would not be altered by a more accurate modeling of ϵ_F nor by a small correction for ξ/l.

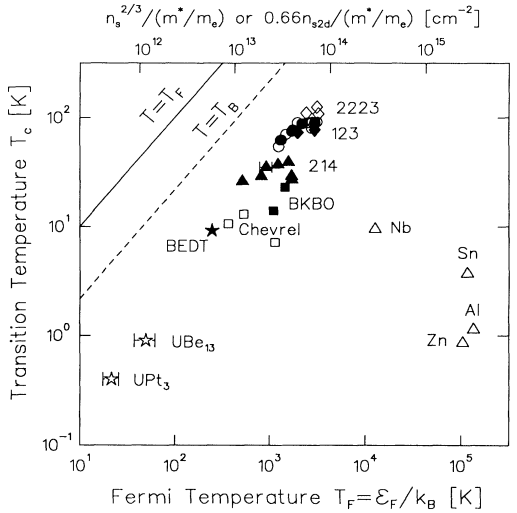

FIG. 3. A log-log plot of T_c vs the Fermi temperature $T_F=\epsilon_F/k_B$ estimated from the results of σ in Fig. 2 (combined with the interplanar distance c_{int} for 2D and the Sommerfeld constant γ for 3D systems). The values of T_F for Sn, Al, and Zn are based on knowledge from other estimates. The dashed line represents the BE condensation temperature T_B of the ideal boson gas with corresponding $n_s^{2/3}/m^*$, the boson mass $2m^*$, and density $n_s/2$.

The low carrier density and/or heavy mass make the Fermi energy very small in these exotic superconductors. As described in Refs. 4 and 18, when ϵ_F becomes comparable to or less than the energy $\hbar\omega_B$ of bosons mediating the superconducting pairing, the pairing interaction is no longer retarded, and one can expect $T_c \propto \epsilon_F$. This is one possible way to account for the linear trend in Figs. 2 and 3.

It is also interesting to explore the relation with Bose-Einstein (BE) condensation. A situation close to local bosons is conceivable in view of short coherence length ξ of these exotic superconductors.[3] There is no BE condensation in purely 2D systems. One can, however, expect BE condensation if a small 3D interaction is present, as discussed by Friedberg and Lee.[19] The BE condensation occurs when the thermal wavelength $\Lambda(T)$ [defined through $k_BT \propto p^2/2m^*$ and $h/\Lambda \sim p$, $\Lambda(T) \propto 1/\sqrt{T}$] becomes comparable to the interparticle separation of local bosons.[19] In an ideal 3D Bose gas, $k_BT_B=(1.04\hbar^2)n_s^{2/3}/m^*$ for the boson density $n_s/2$ and mass $2m^*$. The dashed line in Fig. 3 shows the expected values of T_B for given $n_s^{2/3}/m^*$. This line would also serve as a rough measure of the maximum condensation temperature T_B for quasi-2D systems. We see that T_c's of the exotic superconductors are only about 3–30 times smaller than T_B expected for the ideal Bose gas. Thus, these systems lie close to the BE condensation in a thermodynamic sense.

These exotic superconductors have completely different electronic and crystal structures from one series of compounds to another. Therefore, the microscopic pairing interactions and mechanisms likely differ from series to series. This aspect may be reflected in the symmetry of pairing: cuprate, BKBO, and Chevrel systems show s-wave pairing, while some HF systems exhibit unconventional pairing, and our recent data on BEDT also suggest a possibility of line nodes.[3,11] On the other hand, these exotic systems have various features in common. They all have relatively high H_{c2}, short ξ, and highly correlated electronic structures. The present results in Figs. 2 and 3, together with these features, strongly suggest that these systems possibly share a common condensation mechanism and/or thermodynamic description.

Comparison between T_c and T_B in Fig. 3 indicates that the superconducting transition in these exotic systems occurs around the temperature at which the thermal wavelength $\Lambda(T_c)$ of the pairs is 2–6 times longer than the average interpair separation d_{pair}. This situation interpolates the cases of ordinary BCS superconductors $[\Lambda(T_c)/d_{\text{pair}} \gtrsim 50]$ and of ideal real-space local bosons $[\Lambda(T_B)/d_{\text{pair}} \sim 1]$. We note that T_B of the BE condensation in ideal bosons is determined only by the particle density and mass, regardless of the energy scale $\hbar\omega_B$ of the pairing interaction, as long as $\hbar\omega_B \gg kT_B$. The linear trend seen in Fig. 3 common to various different systems may be related to this feature.

These results encourage development of a theory of superconductivity in low carrier densities and strong pairing interactions which interpolates the BCS theory and the BE condensation. Some efforts along this line have been started.[18-20] We should also note that the saturation and suppression of T_c with increasing n_s/m^*, found in cuprate and Chevrel-phase systems, are yet to be explained by such a new theory.

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