Superconductivity on the border of a spin-gapped Mott insulator: NMR studies of the quasi-two-dimensional organic system $EtMe_3P[Pd(dmit)_2]_2$

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We present ^{13}C-NMR studies of the pressure-temperature phase diagram of the quasi-two-dimensional organic system $EtMe_3P[Pd(dmit)_2]_2$, which has a spin-gapped Mott insulating ground state at ambient pressure. As the system is pressurized, the spin-gapped insulating ground state survives until the insulator-metal Mott transition, just beyond which a superconducting state appears. This demonstrates that the superconducting phase borders the spin-gapped insulating phase. The phase diagram contrasts with those of the large majority of other correlated-electron superconductors in which the superconducting phase borders a magnetically ordered phase. This implies the possibility that the present superconductivity has an exotic origin and nature.

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I. INTRODUCTION

Superconductivity in highly correlated electron systems is one of the most fascinating phenomena in condensed-matter physics. It is observed in various materials, such as high-T_c cuprates, f-electron systems, and organics. The mechanism of the superconductivity in highly correlated electron systems is still an open question, though there is a general consensus that the superconductivity is not mediated by the usual electron-phonon coupling. The superconductivity is typically observed adjacent to magnetically ordered phases, which has determined the direction of current theoretical studies into the mechanism of such superconductivity. For example, magnetic fluctuations have been proposed as candidates for the mechanism because they are naturally expected to exist in phases adjacent to magnetically ordered phases.

Recently, a novel superconducting phase which may not border a magnetically ordered phase has been found[1] in a quasi-two-dimensional organic material, $EtMe_3P[Pd(dmit)_2]_2$ (space group $P2_1/m$).[2] This material is a Mott insulator at ambient pressure and has a nearly regular triangular lattice.[1,3] This lattice is similar to that of the spin-liquid material $EtMe_3Sb[Pd(dmit)_2]_2$,[4,5] and strong frustration effect works also in $EtMe_3P[Pd(dmit)_2]_2$. However, unlike $EtMe_3Sb[Pd(dmit)_2]_2$ with the solid-crossing stacking structure,[6] $EtMe_3P[Pd(dmit)_2]_2$ has the parallel molecular stacking structure,[1,3] where lattice is easy to distort. As a result, $EtMe_3P[Pd(dmit)_2]_2$ undergoes a second-order spin-gap phase transition at 25 K accompanied by lattice dimerization.[3] Recent resistivity[1,7] and susceptibility[8,9] studies for this material report that superconductivity appears under pressure. Therefore, it is possible that the superconducting phase does not border a magnetically ordered phase but rather a nonmagnetic phase.

However, studies of this system under pressure have so far measured only macroscopic physical quantities. There is currently no microscopic evidence that the spin-gapped phase survives under pressure until the appearance of the superconductivity. It remains possible that an antiferromagnetic phase might emerge and border the superconducting phase, as is observed in the phase diagram[10] of the TMTTF/TMTSF (Ref. 11) family.

In this paper, we report the first microscopic study of $EtMe_3P[Pd(dmit)_2]_2$ under pressure using ^{13}C-NMR measurements. We provide clear evidence that the spin-gapped phase borders the superconducting phase without an emerging magnetically ordered phase. This contrasts strikingly with the case of the large majority of other correlated-electron superconductors.

II. EXPERIMENT

We prepared fine single crystals of $EtMe_3P[Pd(dmit)_2]_2$ by an aerial oxidation method. For the ^{13}C-NMR measurements, the carbon atoms at both ends of the $Pd(dmit)_2$ molecules were enriched by ^{13}C isotopes, as shown in Fig. 1.

We performed NMR measurements at ambient pressure and under several pressures applied with a BeCu clamp cell. For all the measurements under pressure, we used the same NMR tank circuit without replacing the coil, pressure cell, and NMR probe. As a result, the quality (Q) factor of the circuit is nearly unchanged through the measurements under different pressures. The Q factor depends only on temperature due to the circuit resistance dependence on temperature.

The NMR measurements were performed at the field of about 7.6 T. The NMR spectra were obtained by the Fourier transformation of the spin-echo signals following the $\pi/2$-π

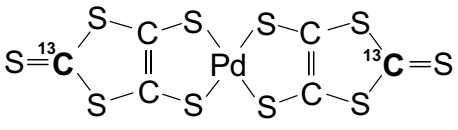

FIG. 1. $Pd(dmit)_2$ molecule with selective substitution of ^{13}C isotope. The carbon atoms at both ends of the molecule are enriched.

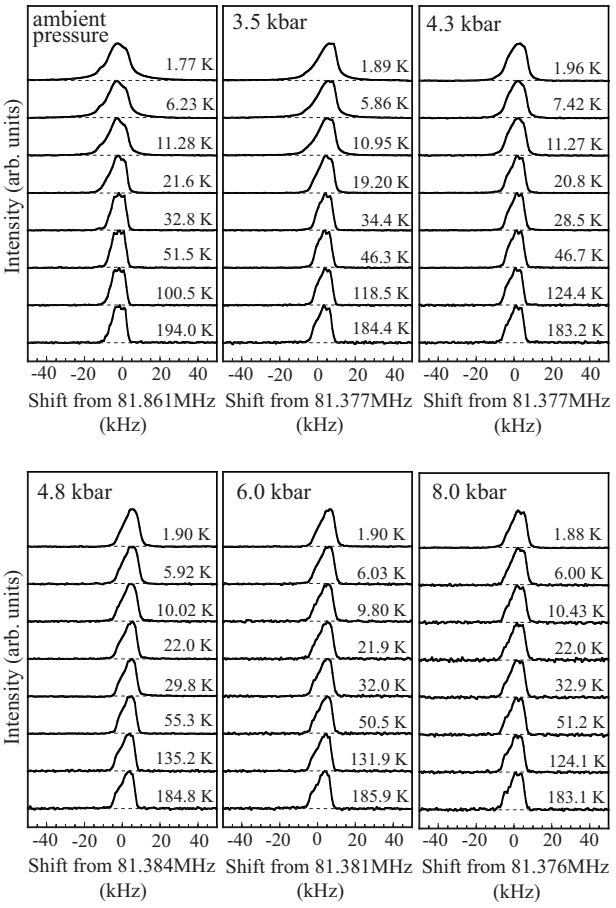

FIG. 2. ^{13}C-NMR spectra for randomly oriented samples of $EtMe_3P[Pd(dmit)_2]_2$ at several pressures.

pulse sequence. The spin-lattice relaxation rate, T_1^{-1}, was obtained from the recovery curve of the integrated spin-echo intensity after irradiating the comb pulses.

We packed a number of fine single crystals into a Teflon tube with no particular orientation. Daphne 7373 oil was used as the pressure medium. Since the natural abundance of ^{13}C isotope is about 1%, the Teflon and Daphne 7373 oil have a slight quantity of ^{13}C atoms. However, due to the fact that they have faster spin-spin relaxation times T_2 than the samples, we succeeded in eliminating signals from the Teflon and oil by setting the interval between the $\pi/2$ pulse and the π pulse to 100 μs, which is much shorter than T_2 of the samples and longer than T_2 of the Teflon and oil.

We applied pressure at room temperature up to 8.0 kbar. The applied pressures were estimated from the external forces applied at room temperature. The estimated values typically have uncertainties of $\pm 5\%$. We note that the pressure at low temperatures decreases by about 1.5–2 kbar from the room-temperature value.[12]

III. RESULTS

Figure 2 shows NMR spectra at various pressures. The displayed pressures are estimated values from the external forces as mentioned in the previous section. The resonance frequencies vary slightly with pressure, though this is not intrinsic but is due to the very slight difference in the applied magnetic fields. All the spectra are sharp and within ± 20 kHz, indicating the absence of magnetic ordering, given that the hyperfine coupling of $X[Pd(dmit)_2]_2$ is about $\sim 9 \times 10^2$ kHz/μ_B.[4]

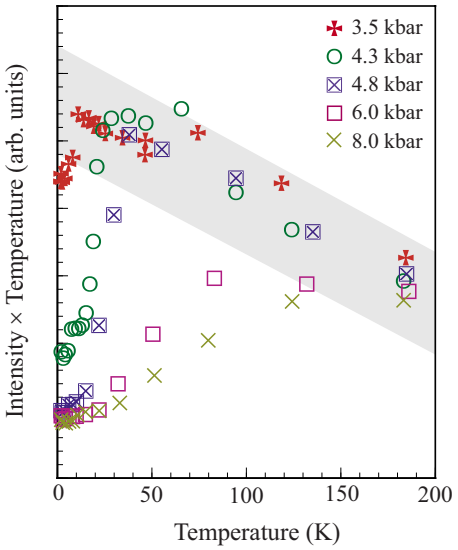

FIG. 3. (Color online) Temperature dependence of the spectral intensity multiplied by temperature. The shaded area indicates expected values when all nuclei contribute to the spectrum without the skin effect.

Figure 3 shows the product of temperature (T) and the intensity of the NMR spectra. The intensity is defined as the integral of the Fourier-transform spectrum over the whole frequency range. It is proportional to both the nuclear magnetization, which varies as $1/T$, and the circuit Q factor. In the present experimental setup, the Q factor increases as the circuit temperature is decreased because the resistance of the circuit decreases. In this case, it is expected that intensity $\times T$ increases monotonically upon cooling along with the increased Q factor, as shown in the shaded area in Fig. 3. However, suppression from this area is observed at low temperatures for high pressures. This is not caused by T_2 decay because T_2 remains almost constant over the entire temperature and pressure region and is approximately 1 ms, which is much larger than the interval between the $\pi/2$ pulse and the π pulse. This suppression indicates that the system becomes highly conductive. When the penetration depth of the electromagnetic field becomes shorter than the sample size due to the growth of the conductivity of the samples, the number of nuclei contributing to the NMR signal, or the NMR intensity, decreases.

The spin-lattice relaxation rate given by stretched-exponential analysis is shown in Fig. 4(a). We have fitted the recovery curves of the nuclear magnetization $M(t)$ by the stretched-exponential function $1-M(t)/M(\infty)=\exp(-t/T_1)^{\beta}$, where β is the stretching exponent and is displayed in Fig. 4(b). The exponent is unity for the single-exponential relaxation but would decrease with increasing distribution of T_1. As shown in Fig. 4(b), the observed exponent deviates from unity even around room temperature. This is because T_1 varies with the angle between the sample direction and applied

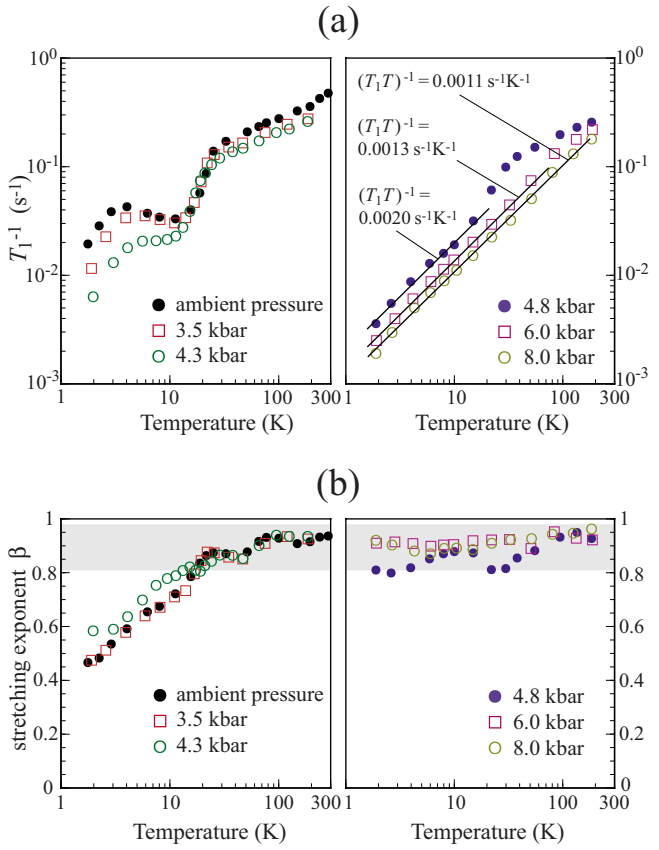

FIG. 4. (Color online) (a) Temperature dependence of ^{13}C nuclear spin-lattice relaxation rate of $EtMe_3P[Pd(dmit)_2]_2$ at several pressures. The rate is determined by stretched-exponential analysis. The solid lines in the right figure are fits to the Korringa relation $(T_1T)^{-1}$=const. (b) Temperature dependence of stretching exponent β at several pressures. The shaded area indicates expected values when the system is homogeneous.

field due to the anisotropy of the hyperfine coupling, and the observed nuclear magnetization M is evaluated as the overall integral of samples with various angles. What is remarkable is a further decrease in β at low temperatures, observed only below 4.3 kbar. This means that the spin system becomes inhomogeneous, which will be discussed in the next section.

Above 4.8 kbar, the measured T_1^{-1} at low temperatures follows the Korringa laws ($T_1^{-1} \propto T$), as are generally observed in conventional Fermi-liquid metals. We should note that the measured T_1^{-1} is reliable, although the skin effect works in this high-pressure and low-temperature region, as mentioned previously. When the skin effect works, a spin diffusion process sometimes affects the nuclear magnetic relaxation and causes a non-single-exponential relaxation curve. However, in the present system, the diffusion process is not considered to work in the time scale of the T_1 measurements because the observed β does not decrease in this high-pressure and low-temperature region, as shown in the right figure of Fig. 4(b). Thus, the effect of the diffusion process is ignorable.

We measured the resonance frequency of the NMR tank circuit under zero magnetic field in order to observe the presence of the superconducting phase. The resonance frequency

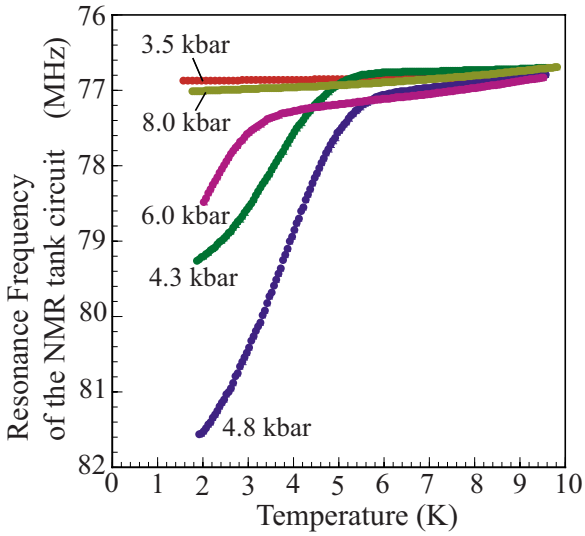

FIG. 5. (Color online) Temperature dependence of the resonance frequency of the NMR tank circuit at zero magnetic field. It reflects the ac susceptibility of the samples. The steep changes in the frequency indicate transitions to the superconducting phase.

is roughly given as $f=1/(2\pi\sqrt{LC})$, where C represents the capacitance of the condenser and L is the inductance of the coil in which the samples are set. A change in L, or a change in the samples' ac susceptibility, is detected by a change in the resonance frequency. The temperature dependences of the resonance frequency at various pressures are shown in Fig. 5. Steep changes in the frequency due to the diamagnetism of the superconductivity are observed for $4.3 \leq P \leq 6.0$ kbar. No hysteretic behavior is observed between the cooling and heating processes within the experimental uncertainty. This diamagnetic behavior was not detected under a magnetic field of 7.6 T, where NMR measurements were performed. Thus, the superconducting states are destroyed in the magnetic field for the present NMR measurements.

IV. DISCUSSION

A. Ambient pressure

We confirmed that there is no magnetic long-range order at ambient pressure. Neither large broadening of the spectra nor critical divergence of T_1^{-1} is observed, as shown in Figs. 2 and 4(a). Instead, T_1^{-1} at ambient pressure shows a rapid decrease below 25 K. This indicates spin-gap formation accompanied by lattice dimerization, which has been proposed in previous susceptibility and x-ray studies.[3]

As is seen in Fig. 4(a), the temperature dependence of T_1^{-1} at ambient pressure below about 10 K shows a positive deviation from a gap-type decrease, suggesting the appearance of another relaxation mechanism. However, this probably does not reflect the bulk magnetic property of the system because the stretching exponent β is far from unity in this region, as shown in Fig. 4(b); i.e., the relaxation is strongly influenced by inhomogeneity. The appearance of such inhomogeneous relaxation is also observed in another lattice-

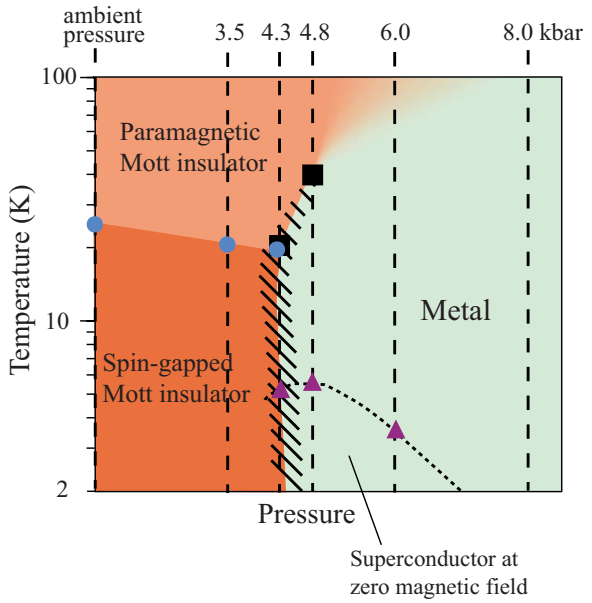

FIG. 6. (Color online) Schematic phase diagram of $EtMe_3P[Pd(dmit)_2]_2$ at 7.6 T. Circles: spin-gap transition temperatures determined by the behavior of T_1^{-1}. Triangles: superconducting transition temperatures at zero magnetic field determined by the onset of the diamagnetic signal. Squares: Mott transition (or rapid crossover) temperatures determined by the decrease in the NMR intensity. According to Ref. 7, the Mott transition is of first order below about 40 K, while it becomes crossover at higher temperatures. The hatched area around the first-order transition indicates the region where the coexistence of the metallic and insulating phases is expected.

dimerized spin-gapped system, $CuGeO_3$.[13] This inhomogeneous relaxation is probably due to remaining unpaired free spins at sites that fail to dimerize. Such defects, if any, are few and thus generally only slightly influence the relaxation. In these spin-gapped phases, however, the spin gap causes the intrinsic relaxation to be so small at low temperatures that the slight relaxation caused by the defects becomes detectable. In this situation, the estimated T_1 by the stretched-exponential analysis shown in Fig. 4(a) reflects not only the intrinsic relaxation but also the extrinsic relaxation due to the remaining free spins. This is probably the reason why the temperature dependence of T_1 at ambient pressure in Fig. 4(a) shows the positive deviation from the gap-type decrease.

In addition, slight broadening of the spectra is observed at ambient pressure below 25 K as shown in Fig. 2, although spin susceptibility vanishes in this region. This is also compatible with the above interpretation that a small number of free spins emerge in the spin-gapped phase.

B. Schematic phase diagram

In this subsection, we determine whether the system at each pressure is in an insulating or metallic phase. The overall result is summarized in Fig. 6.

The suppression of the spectral intensity from the shaded area in Fig. 3 indicates that the system enters a metallic phase. This effect is observed above 4.3 kbar, while it is hardly observed at 3.5 kbar, showing that the low-temperature phase at 3.5 kbar is insulating.[14] At 4.3 kbar, steep suppression of the intensity is observed below around 20 K, which means that the system, or at least a part of the system, becomes highly conductive rapidly due to a metal-insulator transition (or rapid crossover) at this temperature. At 4.8 kbar, the suppression temperature, that is, the transition (or crossover) temperature, rises to about 40 K. At 6.0 and 8.0 kbar, gradual suppressions are observed at higher temperatures, indicating that the conductivity grows gradually from higher temperatures.

In addition, the intensities at the low-temperature limit are the same for 4.8, 6.0, and 8.0 kbar, while the intensity at 4.3 kbar is twice the intensities at these pressures. This implies that the system at 4.3 kbar is not wholly in the metallic phase but includes an insulating phase to some extent.

We show that the metallic phase exhibits an almost single-exponential recovery of the nuclear magnetization, while the spin-gapped insulating phase has an inhomogeneous recovery as discussed in the previous subsection. As shown in Fig. 4(b), the data above 4.8 kbar, where the metallic ground state is realized, do not show a decrease in the exponent β,[15] in contrast to the data at ambient pressure.

At 3.5 kbar, β decreases at the same level as the data at ambient pressure. As for 4.3 kbar, a similar decrease is observed although it is weaker than those at ambient pressure and 3.5 kbar. The decrease in β clearly shows that the system at 4.3 kbar is not wholly in the metallic phase because an almost single-exponential recovery is expected if the system is wholly in the metallic phase. That is, the system at low temperatures for 4.3 kbar includes a bulk insulating phase together with the metallic phase.

From the above intensity and relaxation-curve analyses, the system at low temperatures is in the insulating phase for $P \leq 3.5$ kbar and in the metallic phase for $P \geq 4.8$ kbar. For $P=4.3$ kbar, the intensity analysis and the relaxation-curve analysis demonstrate that the system includes bulk metallic and insulating phases; that is, the metallic and insulating phases coexist at this pressure. This means that the system at 4.3 kbar lies on the Mott transition, as shown in Fig. 6. It has been reported that the Mott transition of the present material is of first order below about 40 K, while it becomes crossover at higher temperatures.[7] Near a first-order transition point, both two phases associated with this transition can exist simultaneously as local-minimum free-energy states, although one of them is a metastable state. This is considered to be the reason why the present system at 4.3 kbar shows the coexistence. We note that, also in the quasi-two-dimensional organic system κ-$(BEDT\text{-}TTF)_2X$,[16] the coexistence of bulk insulating and metallic phases is observed near the first-order Mott transition,[17,18] which is very likely due to the same reason.

The superconducting phase is observed for $4.3 \leq P \leq 6.0$ kbar under zero magnetic field, as shown in Fig. 5. The onset temperature of the superconductivity T_c is 5.0 and 5.5 K at 4.3 and 4.8 kbar, respectively, and reduces to 3.5 K at 6.0 kbar. The magnitude of the diamagnetism at 4.3 kbar is smaller than that at 4.8 kbar, although T_c is almost the same at these two pressures. This is because the system at 4.3 kbar also includes the bulk insulating phase, as discussed above,

and thus is probably not wholly in the superconducting phase.

C. Insulating phase under pressure

The insulating phase under pressure has the same nature as that at ambient pressure. The systems at 3.5 and 4.3 kbar show neither large broadening of the spectra nor critical divergence of T_1^{-1}, as shown in Figs. 2 and 4(a). They show rapid suppression of T_1^{-1} below 20–25 K accompanied by a decrease in β. Below about 10 K, the temperature dependence of T_1^{-1} at 3.5 and 4.3 kbar deviates from a gap-type decrease. This behavior is similar to that at ambient pressure and thus demonstrates that the insulating phase at 3.5 and 4.3 kbar is the same spin-gapped phase as that at ambient pressure. (At 4.3 kbar, a slightly smaller T_1^{-1} and larger β than those at ambient pressure and 3.5 kbar are observed at low temperatures. This is because the data at 4.3 kbar reflect not only the insulating phase but also the metallic phase owing to the coexistence of both phases.)

The spin-gap formation temperature seems to be depressed slightly toward lower temperatures with pressure, as shown in Fig. 4(a).

The system at 4.3 kbar is situated on the boundary of the Mott transition. Thus, our results show that the spin-gapped phase survives an increase in pressure until the Mott transition, not replaced by other phases such as an antiferromagnetically ordered phase. Therefore, it is concluded that the superconducting phase of the present system borders the spin-gapped insulating phase, as shown in Fig. 6.

D. Metallic phase under pressure

The Korringa relation $T_1^{-1} \propto T$ is observed to apply in the metallic phase, as shown in the right figure of Fig. 4(a). This means that the metallic phase has a Fermi-liquid-like low-energy excitation. Although the present metallic phase borders the spin-gapped insulating phase, the legacy of the spin gap, or the pseudo-spin-gap, is not observed in the metallic phase.

Superconductivity is observed only in the vicinity of the Mott transition, as shown in Fig. 6. Thus, we believe that electron correlation plays a key role in the realization of the superconductivity. A strong candidate mechanism of correlated-electron superconductivity on the Fermi liquid is antiferromagnetic fluctuations. In the two-dimensional square-lattice system, enhancement of antiferromagnetic fluctuations at the wave number of (π, π) causes electron pair scattering related to this wave number and, hence, results in the $d_{x^2-y^2}$ superconductivity in appropriate conditions. This idea is also applicable to two-dimensional organic systems with anisotropic triangular lattices, and it is widely thought that the simplest commensurate two-sublattice magnetic fluctuations [which is also abbreviated as (π, π) fluctuations hereafter] cause d-wave-like superconductivity in the same way. This is a strong candidate to explain the mechanism of superconductivity in most of the two-dimensional organic systems such as κ-(BEDT-TTF)$_2X$ because they are considered to border the (π, π) antiferromagnetically ordered phases and thus to have strong (π, π) fluctuations. The metallic phase of the present system, however, does not border the (π, π) ordered phase but rather a spin-gapped phase. This implies that (π, π) fluctuations are less enhanced in the present system. Thus, it is possible that the superconductivity in the present system is not the usual d-wave one mediated by (π, π) fluctuations.

One of the possible scenarios is that the present superconductivity is realized by different wave-number fluctuations, which is likely to be relatively significant in the present frustrated system, where (π, π) fluctuations are depressed. Indeed, it is predicted that exotic superconductivity such as $d+id$ one with broken time-reversal symmetry is realized by the frustrated spin fluctuations on nearly regular triangular lattice.[19]

As another possible scenario, softening dimerization phonons inducing the spin gap might play an important role. It has been proposed theoretically that these phonons can cause s-wave superconductivity.[20,21]

From the viewpoint of a measure of magnetic fluctuations, of interest is the Korringa ratio $K(\alpha)$, which is defined as $(T_1T)^{-1}/(A\chi)^2S$, where A and χ are the hyperfine coupling constant and susceptibility, respectively, and S is $(\gamma_n/\gamma_e)^2(4\pi k_B/\hbar)$. It is unity for noninteracting electrons and increases from unity as antiferromagnetic fluctuations are enhanced by electron correlation. In the present metallic phase, $(T_1T)^{-1}$ is 0.0020, 0.0013, and 0.0011 s^{-1} K^{-1} at 4.8, 6.0, and 8.0 kbar, respectively, as shown in Fig. 4. These values correspond to $K(\alpha)$ of about 2, by taking $A \sim 9 \times 10^2$ kHz/μ_B (Ref. 4) and $\chi \sim 4 \times 10^{-4}$ emu/mol.[9] [More precisely, $K(\alpha)$ is calculated to be 2.6, 1.7, and 1.4 at 4.8, 6.0, and 8.0 kbar, respectively, using the above A and χ values. Strictly speaking, however, χ also decreases with pressure. Thus, it is not clear whether $K(\alpha)$ decreases with pressure or not.] The value $K(\alpha) \sim 2$ is smaller than the reported value[22] of $K(\alpha)=5-10$ for the representative two-dimensional organic superconductors κ-(BEDT-TTF)$_2X$ [X=Cu[N(CN)$_2$]Br and Cu(NCS)$_2$], in which superconducting phases border (π, π) antiferromagnetically ordered phases. This may be evidence that the (π, π) fluctuations in the metallic phase of EtMe$_3$P[Pd(dmit)$_2$]$_2$ are less enhanced than those of κ-(BEDT-TTF)$_2X$. However, we should note that the above values of A and χ have some uncertainties, and thus $K(\alpha)$ for the present material estimated in the above analysis has an uncertainty of a factor of about 2. The Knight shift provides an alternative method to determine $A\chi$ term. Unfortunately, owing to the small hyperfine coupling constant ($A \sim 9 \times 10^2$ kHz/μ_B), the Knight shift of the present material is so small that we cannot estimate the shift precisely. Further study is needed for sufficient discussion of $K(\alpha)$.

V. CONCLUSION

We have presented the pressure-temperature phase diagram of EtMe$_3$P[Pd(dmit)$_2$]$_2$ determined by ^{13}C-NMR measurements. The system is insulating for $P \leq 3.5$ kbar, while a metallic phase is realized at low temperatures for $P \geq 4.8$ kbar. Both these phases coexist at $P=4.3$ kbar. The

Korringa relation is observed to apply in the metallic phase, which means that it has a Fermi-liquid-like low-energy excitation without a pseudo-spin-gap. We provide clear evidence that, in the insulating phase, the spin-gapped ground state survives over the whole pressure region and is not replaced by a magnetically ordered state. Therefore, the superconducting phase of $EtMe_3P[Pd(dmit)_2]_2$ borders the spin-gapped insulating phase, as shown in Fig. 6. This stands in striking contrast to the phase diagram of the large majority of other correlated-electron superconductors and therefore suggests the possibility that the present superconductivity has an exotic origin and nature.

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[14] We observe a very slight decrease in the NMR intensity at 3.5 kbar below about 10 K in Fig. 3. We cannot assess whether the decrease is reliable or within experimental error. Even if it is intrinsic, the volume fraction of the metallic phase is extremely low.

[15] The data at 4.8 kbar tend to have slightly smaller values of β than the data at 6.0 and 8.0 kbar. Although we cannot conclude whether this is reliable or within experimental error, an extremely low volume of the insulating phase possibly exists at 4.8 kbar.

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