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Weak suppression of ferromagnetism in tetrakis(dimethylamino)ethylene-$(C_{60})_{1-x}(C_{70})_x$

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Ferromagnetic properties of the molecular alloy complexes TDAE-$(C_{60})_{1-x}(C_{70})_x$ ($x=0.1-0.9$), where TDAE is tetrakis(dimethylamino)ethylene, are studied by means of ESR and static magnetic susceptibility and magnetization experiments. Comparison of the magnetic properties of the present alloy complexes with those of TDAE-C_{60} and -C_{70} is made. The ESR g values and linewidths of these complexes remarkably increase below the Curie temperature $T_c(x)$ at $x<0.5$. Moreover, these complexes with $x=0.2$, 0.4, and 0.5 show the S-shaped magnetization curves indicative of ferromagnetic correlation of spins below the $T_c(x)$. The $T_c(x)$ is suppressed with increasing x in such a way that $T_c(0)=17.5$ K and $T_c(0.5)=6.1$ K. This suppression is not so strong as one may expect for a quasi-one-dimensional itinerant ferromagnet. The three-dimensional spin correlations between C_{60} molecules both along the c axis and in the ab plane are important for this interesting magnetism.

I. INTRODUCTION

TDAE-C_{60} has been demonstrated to undergo a ferromagnetic transition at 16.1 K and to show magnetization without remanence by Allemand *et al.*,[1] where TDAE is tetrakis(dimethylamino)ethylene, one of the strongest organic electron donors. An x-ray powder diffraction study[2] has shown that the stoichiometry of TDAE-C_{60} is a 1:1 charge-transfer complex and that the structure has c-centered monoclinic unit cell suggestive of an anisotropic band structure. Our group has found by using ESR and Faraday-type magnetic balance that a ferromagnetic transition of TDAE-C_{60} occurs at 16.7–17.5 K.[3,4] The Curie temperature (T_C) of this material is the highest in the molecular ferromagnets[5] made of first-row elements such as C, N, and O hitherto reported. We have observed a remarkable increase in the ESR g value and broadening of the ESR line shape in TDAE-C_{60} below the T_c as well as the S-shaped magnetization curve indicative of a ferromagnetic correlation of spins.[3,4] Our ESR results on TDAE-C_{60} have been reconfirmed.[6] Recently TDAE-C_{60} was reported, on the basis of ESR analyses, to exhibit a ferromagnetic transition at 24 K under special conditions.[7] An evidence for a magnetic hysteresis curve in TDAE-C_{60} was also reported.[8]

Furthermore, we have found that TDAE-higher-fulleride complexes such as TDAE-C_{70}, -C_{84}, -C_{90}, and -C_{96} do not show a ferromagnetic transition but only paramagnetism at least above 4.5 K.[3,4,9] Also we have observed that there is a disproportionation between C_{60}^{-} and C_{60}^{2-} in dilute solution of TDAE-C_{60}, but not in TDAE-C_{70}.[10] From these results, we consider that C_{60} is exceptional in its magnetic properties as well as in superconductivity, which is absent in M_xC_n, where M = K and Rb, and $n=70$, 76, 78, 84, and 90.[11]

Moreover, observation of a strong decrease in the superconducting transition temperature of the molecular alloy complexes $M_x(C_{60})_{1-y}(C_{70})_y$ is of interest.[11,12] This motivated us to investigate the change in the ferromagnetic T_c of the molecular-alloy complexes, TDAE-C_{60} containing C_{70} as impurities. It has been suggested, on the basis of the relatively high electrical conductivity and the crystalline structure analysis,[1,2] that the origin of the ferromagnetic transition in TDAE-C_{60} is the itinerant spins in metallic quasi-one-dimensional (Q-1D) C_{60} chains along the c axis.[1,13] A large pressure dependence of this ferromagnetism has supported this picture.[14] If the Q-1D nature is strong in this material, one may expect that a certain amount of C_{70} as impurity should induce the electrons in the C_{60} chain to localize by the Anderson mechanism[15] and hence readily destroy the itinerant ferromagnetism. On the other hand, the existence of C_{70} would less dramatically induce the disappearance of ferromagnetism if the spin carriers come from the localized spins in a three-dimensional band structure.

We have reported preliminarily that a weak suppression of ferromagnetism occurs in TDAE-C_{60} containing C_{70} as impurity.[16] In this paper we like to report a detailed study of the magnetic properties of TDAE-$(C_{60})_{1-x}(C_{70})_x$, where x ranges from 0.1 to 0.9, with combined analyses of ESR, static magnetic susceptibility, and magnetization measurements. Based on these investigations we further try to clarify the origin of the ferromagnetic property of TDAE-C_{60}.

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II. EXPERIMENTAL PROCEDURES

The as-purchased fullerenes C_{60} and C_{70} (purity: 99.9 and 98.2%, respectively) from Vacuum Metallurgical Co., Ltd. were employed without further purification. TDAE purchased from Tokyo Chemical Industry Co., Ltd. was purified by distillation under vacuum at 30°C. Required amounts of C_{60} and C_{70} were dissolved in a minimum amount of benzene-toluene (1:1) mixed solvent in a glass vessel, and then the solution was degassed by several freeze-pump-thaw cycles. Note that C_{60} and C_{70} are completely soluble in this mixed solvent and that the solutions prepared in this study were nearly saturated. After that, an excess amount of TDAE was added under nitrogen atmosphere and degassing was again performed. Black precipitate of TDAE-$(C_{60})_{1-x}(C_{70})_x$ was dried under dynamic vacuum for 30 hours and then the sample was placed in an ESR tube of o.d. 5 mm under a nitrogen flow and sealed under vacuum. Five kinds of samples of 1 mg with $x=0.1$, 0.3, 0.5, 0.7, and 0.9 were prepared for the ESR measurements. Three samples of 30 mg with $x=0.203$, 0.374, and 0.477 were prepared for static magnetic measurements and these samples are simply referred to as $x=0.2$, 0.4, and 0.5 throughout this paper. Stoichiometry of TDAE and the C_{60}-C_{70} alloys (molar ratio of TDAE/fullerene alloy) could be assumed to be approximately 1 based on the previous results for TDAE-C_{60} and TDAE-C_{70} with TDAE/fullerene being about unity.[1–4]

The crystalline structures of $(C_{60})_{1-x}(C_{70})_x$ molecular alloys depending on the type of preparation method have been studied,[17] where it has been shown that the $(C_{60})_{1-x}(C_{70})_x$ form solid solutions with fcc lattice at $x<0.5$ having a slightly larger lattice constant compared with the pure C_{60} crystal. Hence one may expect that the lattice structure of the present TDAE-$(C_{60})_{1-x}(C_{70})_x$ alloys should not differ much from that of TDAE-C_{60}. Also note that C_{60} and C_{70} are apparently not segregated in the present molecular alloy complexes because of a monotonic and sequential suppression of the Curie temperature as a function of x as described below.

The ESR spectra were recorded on Bruker ESP 300E ESR spectrometer with an Oxford liquid-helium-flow cryostat at the X band with a field modulation of 100 kHz in the temperature range 2.5–270 K. During single ESR measurements, the temperature was maintained within ±0.5 K of its set value. Peak-to-peak linewidth ($\Delta H_{\mathrm{p.p.}}$) and g values were determined using a Mn^{2+}-MgO standard as usual.[3,4] ESR intensities were calculated from double integration of the first derivative ESR spectra and spin concentration was determined by using $CuSO_4 \cdot 5H_2O$ as a standard. The relative errors of the observed $\Delta H_{\mathrm{p.p.}}$ and g values are less than 4% and 0.03%, respectively. The magnetization and the magnetic-susceptibility measurements were carried out with an Oxford Instruments Faraday-type magnetic balance. The sample sealed in a quartz ESR tube of 30 mm in length was hung in the magnetic field using a very thin quartz fiber. Magnetic susceptibility of the ESR tube was calibrated from the raw data using standard methods.[4] The magnetization curves were recorded at 4.5 and 19.5 K under magnetic field change $0 \rightarrow 54$ kG $\rightarrow 0 \rightarrow -54$ kG $\rightarrow 0 \rightarrow 54$ kG $\rightarrow 0$. The temperature dependence of the magnetic susceptibility was examined in the range 2.4–253 K at the constant magnetic field of 5 kG.

III. RESULTS AND DISCUSSION

A. ESR analysis

The g values of TDAE-$(C_{60})_{1-x}(C_{70})_x$ of $x=0.1$, 0.3, 0.5, 0.7, and 0.9 are plotted as a function of temperature in Fig. 1. We have reported that the g value of TDAE-C_{60} is 2.0003 at room temperature while that of TDAE-C_{70} is 2.0022 and that the line shape of TDAE-C_{60} becomes asymmetric below 40 K.[3,4] The temperature dependence of the g values in TDAE-$(C_{60})_{1-x}(C_{70})_x$ is found to depend on x, the fractional content of C_{70}. This result confirms that C_{60} and C_{70} are homogeneously mixed in these samples. A large increase in the g value was observed below T_c at $x<0.5$ while the g value change is small for $x>0.7$. The signal of $TDAE^{+}$ could not be observed in TDAE-$(C_{60})_{1-x}(C_{70})_x$, which is consistent with our previous results on TDAE-C_{60} and -C_{70}.[4] Hence, we consider that the magnetism of these materials comes from the fullerene component and that the spins of $TDAE^{+}$ are cancelled in the crystalline structure or, at least, in the bulk.

Figure 2 shows the temperature dependence of $\Delta H_{\mathrm{p.p.}}$ of TDAE-$(C_{60})_{1-x}(C_{70})_x$. $\Delta H_{\mathrm{p.p.}}$ of all the samples become narrower with decreasing temperature above T_c. We have attributed this narrowing to the increase in exchange interaction among electronic spins in the previous paper.[3,4] However, since the narrowing at low temperatures has been observed both in solid[3,4,18–20] and in dilute

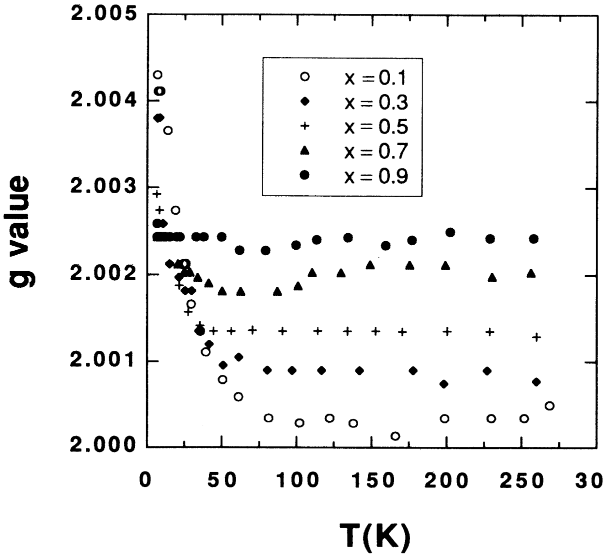

FIG. 1. Temperature dependence of g values of TDAE-$(C_{60})_{1-x}(C_{70})_x$.

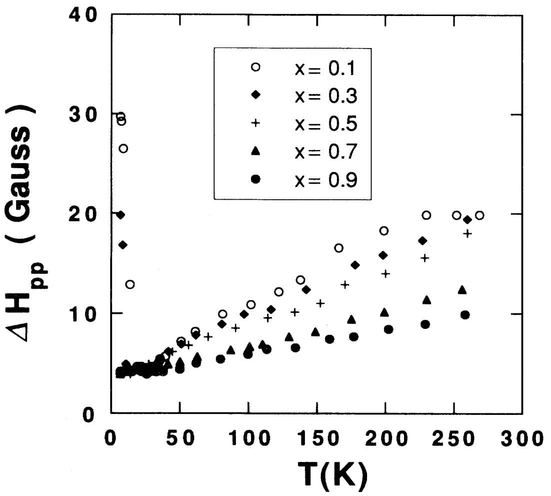

FIG. 2. Peak-to-peak linewidth ($\Delta H_{\mathrm{p.p.}}$) of TDAE-$(C_{60})_{1-x}(C_{70})_x$ as a function of temperature.

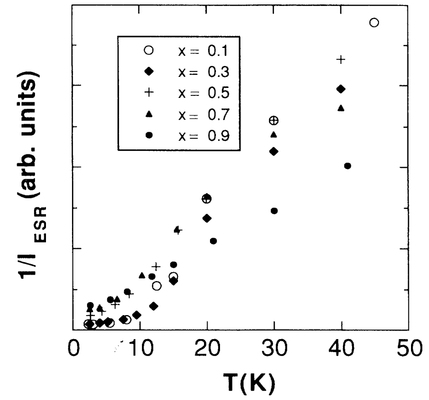

FIG. 3. Inverse ESR intensity ($1/I_{\mathrm{ESR}}$) of TDAE-$(C_{60})_{1-x}(C_{70})_x$ as a function of temperature.

solution,[8,21–23] this phenomenon would be further attributable to the intrinsic property of a C_{60}^- molecule itself. Hence the change in $\Delta H_{\mathrm{p.p.}}$ upon the temperature in TDAE-C_{60} is considered to be similar to that of a C_{60}^- molecule[21,23] for the following reason: The geometry of C_{60}^- is slightly deformed from I_h symmetry into D_{3d}, D_{5d}, or D_{2h} through the Jahn-Teller distortion.[24,26] Since the energy difference of these three structures is quite small (~ 0.02 kcal/mol),[25] the line broadening at high temperatures is considered to be mainly due to the averaging of the Jahn-Teller distorted structures by pseudo-rotation.[23] Note that the degree of broadening is found to be larger in smaller x values, that is, in C_{60}-richer materials as seen in Fig. 2.

On the other hand, C_{70} has no degeneracy both in the highest occupied molecular orbital (HOMO) and in the lowest unoccupied molecular orbital (LUMO)[27] and hence, C_{70}^- should have spin-only paramagnetism.[28] As a result, the g value and $\Delta H_{\mathrm{p.p.}}$ of TDAE-C_{70} are similar to those of conventional organic radicals[29] and almost independent of temperature, in remarkable contrast to those of TDAE-C_{60}. The g values and $\Delta H_{\mathrm{p.p.}}$ of the present molecular alloys are thus consistent with the results of TDAE-C_{60} and TDAE-C_{70}. $\Delta H_{\mathrm{p.p.}}$ is remarkably increased again below T_c at $x<0.7$ concomitant with the temperature dependence of the g values. These phenomena can be clearly ascribed to the appearance of ferromagnetic correlation of TDAE-$(C_{60})_{1-x}(C_{70})_x$.

The spin concentrations at $x=0.1$, 0.3, 0.5, 0.7, and 0.9 were determined to be 5×10^{23}, 5×10^{23}, 4×10^{23}, 3×10^{23}, and 3×10^{23} per gram, respectively. Although all the spin concentrations are of the order of the Avogadro number, they decrease very slightly with increasing the C_{70} content. The inverse ESR intensities ($1/I_{\mathrm{ESR}}$) of TDAE-$(C_{60})_{1-x}(C_{70})_x$ are shown as a function of temperature in Fig. 3. These curves are found not to obey the Curie-type behavior due to the ferromagnetic interactions. T_c can be estimated from these curves. The change in T_c of TDAE-$(C_{60})_{1-x}(C_{70})_x$ will be described together with the results of static magnetic measurements in the following section.

B. Magnetization and magnetic susceptibility

The magnetization (M-H) curves at 4.5 and 19.5 K of the present samples of $x=0.2$, 0.4, and 0.5 are shown in Figs. 4(a) and 4(b), respectively. These figures show that the magnetization of these samples tends to saturate at 4.5 K and not at all at 19.5 K, suggesting that the ferromagnetic transition occurs in all the samples in the range 4.5–19.5 K. The temperature dependence of the inverse magnetic susceptibility ($1/\chi$) in Fig. 5 shows similar behavior to that of TDAE-C_{60},[3,4] which allows us to estimate the Curie temperature for all these samples. It is obvious that T_c is suppressed with the increase in the amount of C_{70} in the present samples. This decrease resembles that of T_c of superconductivity in $M_x(C_{60})_{1-y}(C_{70})_y$ (M = K, Rb).[9,10] General suppression of superconductivity has been discussed based on the BCS-type relation.[30] On the other hand, the χ^{-1}-T behavior of all the present samples in Fig. 5 indicates the Curie-Weiss relation with an antiferromagnetic interaction at high temperatures and the ferromagnetic state below each Curie temperature, which is quite similar to TDAE-C_{60}.[3,4] It should be noted here that the change of the Weiss constant Θ from -57 K at $x=0$[4] apparently takes place but cannot be estimated straightforwardly due to the deviation from simple Curie-Weiss behavior in the present molecular alloy as seen from nonlinear dependence of χ^{-1}-T in Fig. 5. The temperature-independent susceptibility also suggests the possible presence of Pauli-paramagnetic behavior due to intermolecular polaronic bands[24] originated from C_{60}^- in the complex.

The values of T_c of TDAE-$(C_{60})_{1-x}(C_{70})_x$ are plotted as a function of x in Fig. 6, where the open circles and filled diamonds are those obtained from the Faraday method and ESR, respectively. An important observation in Fig. 6 is the surprisingly weak suppression of ferromagnetism and the nearly linear dependence of T_c on

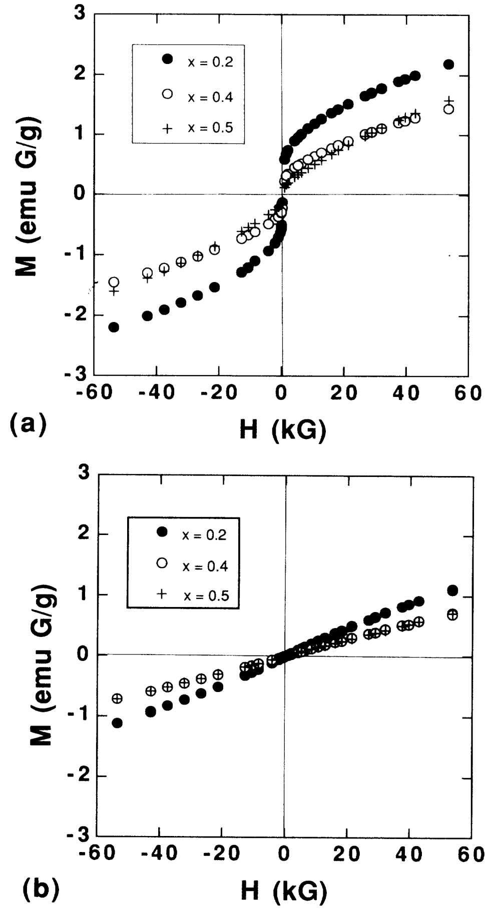

FIG. 4. Magnetization curves of TDAE-$(C_{60})_{1-x}(C_{70})_x$ at (a) 4.5 K and (b) 19.5 K.

x. Note that, moreover, this suppression is less dramatic than that of superconductivity in $K_3(C_{60})_{1-x}(C_{70})_x$.[9,10] That is, superconductivity is not observed already at $x=0.3$ at least above 4.5 K, while the ferromagnetic T_c in TDAE-$(C_{60})_{1-x}(C_{70})_x$ is still comparatively high (6.1 K) even at $x=0.5$.

On the basis of the results described above, the suppression of this ferromagnetism due to C_{70} is found to be very weak. We therefore do not consider that TDAE-C_{60} is a Q-1D system, because in fact the first nearest neighboring distance between C_{60} molecules is 9.98 Å along the c axis and the second nearest is 10.28 Å in the ab plane.[2] If the Q-1D properties of this ferromagnetism are significant along the c axis as mentioned above, the C_{60} metallic chain should be interrupted at low x on the assumption that substitution of C_{60} with C_{70} takes place uniformly. Moreover, as a result, the ferromagnetic properties would disappear because the exchange interaction does not work through the bulk in such a disordered Q-1D system. However, our experimental results exhibit that the ferromagnetism of TDAE-$(C_{60})_{1-x}(C_{70})_x$ survives even at sufficiently large x, for instance, at $x=0.9$.

The difference in the length of the largest diameters of C_{60}^{-} and C_{70}^{-} has been estimated to be 9.3% to that of C_{60}^{-}, being not so much large[31] and, hence, we regard that the lattice structure of TDAE-$(C_{60})_{1-x}(C_{70})_x$ is not remarkably different from that of TDAE-C_{60} at small x. The electronic structure of C_{70} is, however, rather different from that of C_{60} due to the lower symmetry of C_{70}.[27] We

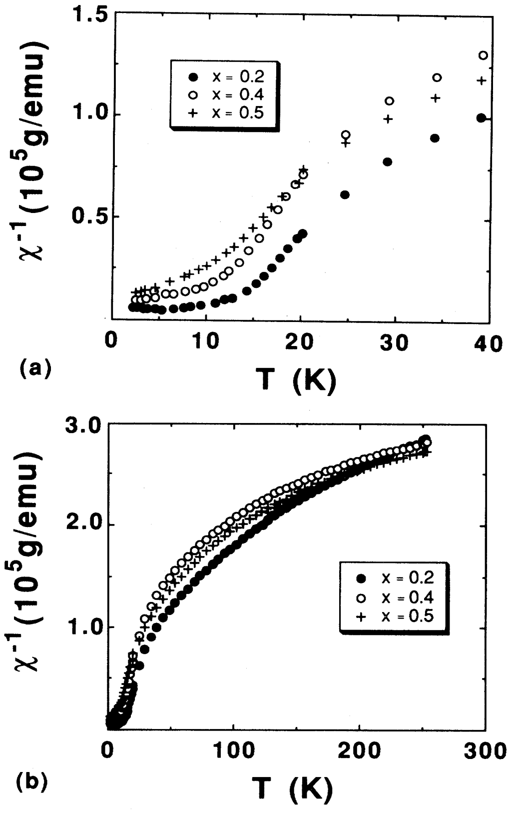

FIG. 5. χ^{-1}-T plot for TDAE-$(C_{60})_{1-x}(C_{70})_x$ in the temperature range, (a) 0–40 K and (b) 0–300 K.

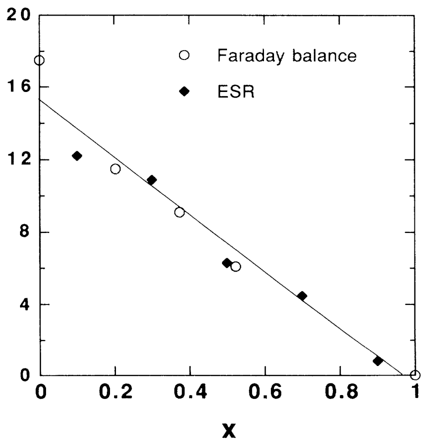

FIG. 6. Correlation between T_c and x of TDAE-$(C_{60})_{1-x}(C_{70})_x$. The open circles were obtained from the static magnetic measurements and the filled diamonds from the ESR analyses of Fig. 3, and the line was based on Eq. (2). T_c was tentatively set 0 for TDAE-C_{70}.

hence consider that C_{70} molecules should easily induce the Anderson localization[13] as impurities in the Q-1D bands along the c direction even at very small x. Although the crystalline structures of TDAE-$(C_{60})_{1-x}(C_{70})_x$ and their electrical conductivity change with x should be studied to understand the origin of this ferromagnetism, the present weak-T_c suppression is a rather unexpected result in the context of the previous observation of the superconductivity in $K_3(C_{60})_{1-x}(C_{70})_x$.[9,10]

On the other hand, the observed decrease in ferromagnetic $T_c(x)$ can be qualitatively discussed from the mean-field theory[32] on the assumption of spin-only magnetism,[28]

$$T_c = 2JzS(S+1)/3k_B \ , \tag{1}$$

where J is the exchange integral, z the number of the nearest neighbors, S the spin quantum number on the central site, and k_B the Boltzmann constant. Since it has been reported that TDAE-C_{60} has a c-centered monoclinic unit cell,[2] considering the first and second nearest interactions, the central C_{60} molecule is surrounded by six neighbors, i.e., $z=6$, as illustrated in Fig. 7(a) assuming $J_1 \approx J_2 \equiv J$, where J_1 and J_2 correspond to the first and second nearest interactions. On close inspection of the ESR results,[3,4,8] it has been found that TDAE does not explicitly contribute to the magnetic properties of both TDAE-C_{60} and TDAE-C_{70} solids. As shown in Fig. 7(b), if one supposes that C_{70} molecules substitute at nearest neighboring sites in proportion to x in TDAE-$(C_{60})_{1-x}(C_{70})_x$, Eq. (1) can be modified into

$$T_c(x) = 4J(1-x)S(S+1)/k_B \tag{2}$$

if one assumes $J_3=0$, where J_3 is the exchange integral between C_{60} and C_{70}. Since S is $\frac{1}{2}$ on a C_{60} molecule from the ESR measurement for TDAE-C_{60}, the exchange integral J is estimated to be 5.03×10^{-4} eV ($= 4.05$ cm^{-1}), being smaller than the reported value of 50 K ($=34.8$ cm^{-1})[6] by about one order of magnitude.

The T_c values estimated from Eq. (2) are also shown in Fig. 6. The present experimental values agree well with this line, signifying that the mean-field theory in Eq. (1) is qualitatively applicable to the present TDAE-$(C_{60})_{1-x}(C_{70})_x$ molecular alloy complexes. This fairly plausible relationship seems to support that the exchange integral works only between C_{60} molecules in the TDAE complex and that between C_{60} and C_{70} molecules vanishes, which agrees with TDAE-C_{70} showing the complete Curie paramagnetism above 1.8 K with zero Weiss constant. Note that the above assumption of zero exchange integral between C_{60} and C_{70} ($J_3=0$) could be justified from their different LUMO levels in the McConnell model for charge-transfer complexes[33] or its modification.[11] It has been reported from molecular-orbital calculations that in C_{70}^- there is a spin-polarized distribution occurring from an excessive spin[27] in contrast to C_{60}^- possessing the "polaron belt" around its central or "equator" area of the sphere.[24] The present experimental results are also consistent with such differences in the electronic structures of C_{60}^- and C_{70}^- molecules that could additionally contribute to the intermolecular exchange integral.

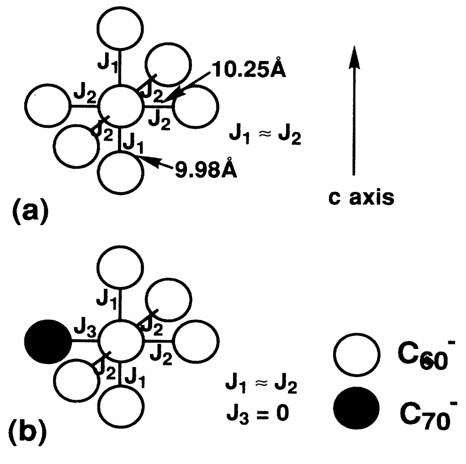

FIG. 7. Schematic representation around a C_{60} molecule in TDAE-$(C_{60})_{1-x}(C_{70})_x$ with (a) $x=0$ and $x=\frac{1}{6}$.

IV. SUMMARY AND CONCLUSIONS

We have investigated the suppression of the ferromagnetism of TDAE-C_{60} containing C_{70} as impurities by means of the ESR and the Faraday method. There are three major findings in the present study, as follows.

(i) The $T_c(x)$ of TDAE-$(C_{60})_{1-x}(C_{70})_x$ decreases monotonically with x in such a way $T_c(0)=17.5$ K and $T_c(0.5)=6.1$ K. This suppression was extremely weak in contrast to that of the superconductivity in $K_3(C_{60})_{1-x}(C_{70})_x$.

(ii) The temperature dependence of the ferromagnetic $T_c(x)$ values agrees well with the prediction from the mean-field theory, where we consider the first and the second nearest interactions, on the assumption that the exchange integral between C_{60}^- and C_{70}^- is zero. This simple model is qualitatively applicable to the present magnetism in the whole range of x as shown in Fig. 6.

(iii) The present study suggests that the ferromagnetism of TDAE-C_{60} is attributable to the three-dimensional interactions among C_{60}^- molecules, because if TDAE-C_{60} is a Q-1D system, the exchange interaction would not work through the bulk even at low x. The interactions both along the c axis and in the ab plane in this molecular crystal would be important for the realization of this interesting ferromagnetism.

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