Metal-to-insulator evolution in \((\text{NH}_3)_x\text{NaK}_2\text{C}_{60}\): an NMR study

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A singular evolution toward an insulating phase, shown by \(^{23}\text{Na}\) and \(^{13}\text{C}\) NMR, has been observed in the superconducting fullerides \((\text{NH}_3)_x\text{NaK}_2\text{C}_{60}\) for \(x > 1\). Unlike most common cases, this insulating phase is non magnetic and \(^{13}\text{C}\) spin lattice relaxation shows the presence of a spin gap. These two features suggest that a charge disproportion from \(\text{C}^{2+}\) to \(\text{C}^{4+}\) can drive the system from the metallic to the insulating state.

The restoring of the \(\text{Na}^+\) cation in the center of the octahedral interstice in the insulating phase, as indicated by \(^{23}\text{Na}\) and \(^2\text{H}\) lineshape analysis, confute the current belief that the cation off-centering is effective in quenching the superconductivity.

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I. INTRODUCTION

Soon after the discovery of fullerene it was found that compounds of the \(\text{A}_x\text{C}_{60}\) family, obtained by insertion of alkali metals \((\text{A})\) in the \(\text{C}_{60}\) lattice, exhibited superconducting behaviour with high critical temperatures \(T_c\) (up to \(\sim 30\) K). A regular increase in transition temperatures was observed as the cation radius and lattice parameter increased, a trend considered nowadays as a textbook example of the success of standard BCS theory. Indeed, according to this theory, \(T_c\) is strongly dependent upon the density of states at Fermi level \(\rho(E_F)\), a quantity which, as a rule, increases with the band-narrowing caused by lattice expansion. This mechanism for increasing \(T_c\) proved rather successful, so attempts to find alternative “lattice expanders” soon followed, thus suggesting the neutral molecule of ammonia as a good candidate. A well known example is given by \(\text{Na}_2\text{CsC}_{60}\). After ammoniation, it becomes \((\text{NH}_3)_x\text{Na}_2\text{CsC}_{60}\), with an increase in transition temperature from 10.5 K to 29.6 K. In general, a further narrowing of the band is expected to enhance the electron correlation, which eventually induces a Mott-Hubbard transition to an insulating magnetic phase. This is just the case of \(\text{K}_2\text{C}_{60}\), \(\text{K}_2\text{RbC}_{60}\) and \(\text{Rb}_2\text{C}_{60}\), which after ammoniation become insulating and antiferromagnetic. In particular the superconductivity of \(\text{NH}_3\text{K}_2\text{C}_{60}\) is recovered after application of external pressure.

An exception to this simple picture is given by the \((\text{NH}_3)_x\text{NaK}_2\text{C}_{60}\) and \((\text{NH}_3)_x\text{NaRb}_2\text{C}_{60}\) compounds. Here the progressive increase of ammonia concentration \(x\) yields an increase in lattice parameter and an unexpected decrease of \(T_c\). This anomalous behaviour was originally attributed to the octahedral cation off-centerings. Indeed, in these systems the \(\text{NH}_3\)-Na groups occupy the large octahedral \((\text{O})\) sites of the \(\text{fcc}\) lattice and the breaking of the cubic symmetry of the crystal field could lift the degeneracy of the \(\text{C}_{60} \Gamma_{1_u}\) LUMO, thus inducing a bandwidth increase and a corresponding decrease in \(\rho(E_F)\). In a recent work we have found that in \((\text{NH}_3)_x\text{NaK}_2\text{C}_{60}\) with \(0.5 < x < 0.9\) the \(\rho(E_F)\), as extracted from Pauli spin susceptibility, actually increases on increasing \(x\) and the lattice parameter, thus resulting inversely correlated with \(T_c\) (anti-Migdal behaviour).

II. SAMPLE PREPARATION AND CHARACTERIZATION

The \((\text{NH}_3)_x\text{NaK}_2\text{C}_{60}\) samples were prepared following the procedure outlined in Ref. 12. However, to achieve higher \(x\) values, the final vacuum drying from the ammo-
nia solution was performed at lower temperatures (down to $-23^\circ$C). The accurate ammonia concentration $x$ was extracted from the hydrogen NMR line intensity. This procedure, besides being quite reliable, was consistent also with volumetric methods which, on the other hand, require the destruction of the sample.

Laboratory x-ray powder diffraction confirms the known fcc structure for the compounds with $x < 1$ and, on the other hand, it does not show any change in the structure on increasing ammonia doping, apart from a slight increase in lattice parameter $a$ from 14.39 Å for $x = 0.9$ to 14.44 Å for $x = 2$.

SQUID magnetometry shows a progressive decrease in $T_c$ from 15 K to 9.5 K for ammonia doping in the $0.5 < x < 1$ range. Above $x = 1$ (corresponding to the lowest transition temperature 9.5 K) up to $x = 2$, no further decrease in $T_c$ takes place, but instead a progressive decrease of the shielding fraction was observed, consistent with the gradual formation of a non-superconducting phase. No magnetism was detected in this range.

As mentioned in the introduction, a similar behavior has been already observed in $K_3C_{60}$, $K_2RbC_{60}$ and $Rb_3C_{60}$ which, upon ammoniation, show a Mott transition to an insulating magnetic phase. In our case, however, the absence of magnetism and the modest lattice expansion suggest that other factors may play a role in suppressing the (super)conducting state.

III. $^{23}$Na NMR

Nuclear Magnetic Resonance has proved a valuable tool for the study of the electronic properties of fullerides since the early time of their discovery. Its usefulness mainly arises from the sizeable hyperfine interaction of conduction electrons both with carbon and alkali nuclei. In addition, microscopic structural information can be obtained also from the nuclear quadrupole interactions with electric field gradients (EFG). The latter interaction, however, affects only the resonance of nuclear species with spin $I > 1/2$, so the possible candidates in our case are Na and K. We focused our attention on $^{23}$Na nucleus because, thanks to its octahedral position: (i) a large hyperfine interaction with conduction electrons is expected, (ii) its off-centered position implies the presence of a considerable electric field gradient on it.

Figure 1 shows the $^{23}$Na NMR spectra taken at RT in samples with different ammonia contents in an external field of 6.88 T; the measured resonances comprise all the three transitions, $^{23}$Na being an $I = 3/2$ nucleus (see further).

Let us first consider the shifts of the lines with respect to a reference NaCl aqueous solution. We notice that, as long as the samples are superconducting, we measure a large paramagnetic shift (165 ppm), which is attributed to the spin polarization of the conduction electrons (Knight shift). The shift and the shape of the resonance are consistent with an isotropic (Fermi) hyperfine interaction as expected by the closed shell electronic structure of the ion. The metallic character of the samples with $x < 1$ is not a surprise (this phase having been extensively investigated in previous works). However, upon increasing the ammonia nominal concentration above $x = 1$, a peak at the reference $^{23}$NaCl resonance frequency progressively appears and it becomes the dominant one near $x = 2$, representative of the $^{23}$Na behaviour in an insulating phase.

The residual paramagnetic shift, which is evident in the $x > 1$ spectra of Figure 1, could in principle suggest that the observed 150 ppm shift may be attributed to a (big) change in hyperfine interaction with conduction electrons from the $x < 1$ phase to the $x = 2$ phase and not to a localization of the carriers induced by the developing of an insulating phase. However, the same displacement of the $^{23}$Na line was observed also in superconducting samples ($x < 1$) which were exposed to a weakly oxidizing atmosphere ($O_2 \sim 1000$ ppm). This treatment suppressed the (super)conductivity of the sample without perturbing its structure (no change in the diffraction pattern). This observation supports therefore the suggestion that the disappearance of the paramagnetic shift is indicative of an insulating phase and the residual shift can be attributed to a chemical shift component. As we will see below, also $^{13}$C NMR results will further confirm this issue.

It is important to notice that this system shows two different regimes with respect to ammonia doping: (i) in the range $0.5 < x < 1$ it behaves as a homogeneous solid solution, irrespective of the intrinsic disorder due to the non-integer stoichiometry; this is guaranteed by the fact that differently doped systems behave like homogeneous superconductors with a well defined, $x$-dependent transition temperature; (ii) above $x = 1$ the observation of two coexisting phases (one superconducting and the
other insulating) indicates, on the other hand, that the separation of the two differently doped phases is energetically favourable with respect to the formation of a solid solution. The transition temperature of the residual superconducting phase, observed for \( x > 1 \), identifies its stoichiometry as \( x = 1 \), while its disappearance as \( x \) approaches 2 suggests the emergence of an insulating phase with \( x = 2 \).

Laboratory x-ray powder diffraction shows only cubic reflections in the whole range \( 0.5 < x < 2 \), while a further increase of ammonia doping (above \( x \simeq 2 \)) results in the appearance of tetragonal reflections, which indicate the formation of a different, ammonia richer phase. To avoid contamination with this other (also insulating) phase, we performed our measurements on the sample with \( x = 1.9 \). We would like to stress that the presence of a minority superconducting phase, evident also from Fig. 1 (and probably due to ammonia de-intercalation during the preparation/manipulation of the samples) does not affect the results obtained in this work, since any possible contribution coming from this phase in the measured spectra has been properly identified and taken into account.

As mentioned before, the quadrupolar interaction of the Na nuclei with the electric field gradient is expected to considerably broaden or split the observed spectra, so that a comparison of the resonance line-widths and structure turns out to be very illuminating for the identification of Na\(^+\) ion sites in the two phases. With reference to the left side of Fig. 2 when \( x < 1 \) the width at 290 K is surprisingly small, if we recall that Na\(^+\) ions are assumed to occupy off-center positions in the octahedral interstices, where the local electric field gradients are quite large. Even if we assume that the observed line corresponds to the central \(-\frac{1}{2} \leftrightarrow \frac{1}{2}\) transition, the expected second order quadrupolar broadening would be much greater than the observed one\(^{14}\). We attribute the observation of a narrow line to a rotational narrowing process. Indeed, thanks to the traceless nature of the quadrupolar interaction, an isotropic and fast rotational motion would be effective in averaging it out. The broadening of the line will be, in this case, restored only by freezing the rotational dynamics upon lowering the sample temperature. The temperature dependence of the \(^{23}\text{Na}\) resonance line in the two cases: \( x = 0.6 \) and \( x = 2 \) (superconducting and insulating phase respectively) is shown in Fig. 2.

In the metallic phase the expected broadening is progressively reached on lowering the temperature. At the base temperature \((T = 15 \text{ K})\), only the \(-\frac{1}{2} \leftrightarrow \frac{1}{2}\) transition (affected only at the second order by the quadrupolar interaction\(^{14}\)) survives, while the other two \((-\frac{1}{2} \leftrightarrow -\frac{3}{2}\) and \(\frac{1}{2} \leftrightarrow -\frac{3}{2}\) directly affected by the quadrupolar interaction) have moved too far in frequency to be observed. Further evidence supports this interpretation: (i) the different lengths of the 90 degree pulse at high and low temperatures: a 50% reduction is found in the latter case, where only the central transition is observed\(^{14}\), (ii) the lineshape at low temperature can be fitted quite well using a second order quadrupolar broadening, as shown in the lowest spectrum of Fig. 2. A careful modelling of the observed lineshape at different applied fields\(^{15}\) yields an estimate of 2.19 Å for the off-centering along the [111] direction of the Na\(^+\) ion, in complete agreement with recent diffraction studies\(^{16}\).

All these findings confirm that a motional narrowing process, rather effective in averaging to zero the traceless quadrupolar interaction, takes place at high temperatures. The small bump observed at reference frequency in the room temperature spectrum is assigned instead to the central transition of the remaining \((1-x)\) fraction of Na\(^+\) ions, located in tetrahedral positions. It appears unshifted due to the smaller hyperfine interaction of these ions\(^{14}\), whereas its rather large frequency spread could arise from the static quadrupolar interaction (even at room temperature) of the tetrahedral Na\(^+\) ions with the EFG created by the neighbouring C\(_{60}\) units.

Let’s now turn to the insulating phase (shown in the right side of Fig. 2). In this case the \(^{23}\text{Na}\) resonance line is both unshifted and unbroadered at all the investigated temperatures (only at \( \approx 25 \text{ K} \) a small broadening appears, probably due to the slowing down of the lattice vibrational dynamics and of C\(_{60}\) rotation). If a quadrupolar interaction were present, it would be small and it would induce a weak second order broadening of the central \(-\frac{1}{2} \leftrightarrow \frac{1}{2}\) line, in addition to possible satellite transitions. To ascertain such quadrupolar effects, the detection bandwidth was extended to \( \pm 500 \text{ kHz} \) (a sufficient upper limit, considering the observed peak width). However, no satellites were detected, so we conclude that the narrow resonance is due to a vanishing quadrupolar interaction which merges all transitions in a single reso-
nance line.

To further support this conclusion we performed also an accurate measurement of the 90 degree pulse length, which would be reduced by a factor $\frac{1}{2}$, if only the central transition were irradiated. The pulse length and power for optimal excitation were found to be the same as in NaCl solution, where a single line comprises all the transitions. Moreover, we checked if this condition was still valid when the power was reduced and the pulse length extended, such as to irradiate only a narrow band around the peak. Since no reduction of the effective 90 degree pulse length, with respect to the NaCl solution, was noticed, we confirm that the observed resonance comprises all the transitions thus indicating a vanishing quadrupolar interaction.

The absence of any measurable electric field gradient, in spite of the large $^{23}$Na quadrupole moment ($Q_{Na} \approx 0.11 \times 10^{-28}$ m$^2$), is exactly what we expect in the centre of an environment with cubic symmetry.

In conclusion we find that: a) unlike the superconducting phase, in the insulating one the Na$^+$ cation is located in the centre of the octahedral site and, b) the fcc structure evidenced by diffraction is confirmed.

These findings rise some doubts on the current belief, which associates the decrease in $T_c$ to the cation off-centering. In fact, according to that hypothesis, we would still find a superconducting phase at $x = 2$ with a sizeable increase in $T_c$ instead of the observed insulator.

IV. $^2$H NMR

As a further test and for a better understanding of the motional narrowing mechanism evidenced by $^{23}$Na NMR, we extended our investigation to samples prepared using deuterated ammonia, which allow to carry out $^2$H NMR. Deuterium resonance ($I_{dH} = 1$) is dominated by the quadrupolar interaction with the electric field gradient which, in this case, originates from the $\sigma$ bond with the nitrogen. Thus, the powder lineshape expected for a uniform distribution of ammonia orientations is the typical Pake doublet shown in the lower part of Fig. 3. The expected width of 245 kHz (defined as the distance between singularities) is however reduced by the uniaxial rotation of ammonia around its ternary axis, a mechanism we have found to be effective down to 1.6 K. This dynamics is known to produce an overall reduction of the line width by a factor of $\sim 4.5$ without affecting the spectrum shape. On the other hand, the reorientation of the ternary axis itself can be quite efficient in averaging out the residual traceless quadrupolar interaction, thus making the powder pattern collapse into a single line, as shown in the upper part of Fig. 3 for $x = 0.6$ (superconducting phase), while no narrowing was observed for $x = 2$ (insulating phase).

These results can be rationalized as follows: in the superconducting phase at high temperatures the NH$_3$-Na clusters rotate around the centre of the octahedron thus averaging to zero the quadrupolar interactions of both $^{23}$Na and $^2$H. Upon cooling down from RT this dynamics slows down and the situation appears static at $T \approx 55$ K on the NMR timescale, i.e. the correlation times are expected to be larger than 0.2 ms. The presence of two ammonia molecules in the insulator ($x = 2$ case) inhibits these rotations, which appear to be frozen already at RT.

Since the $^{23}$Na NMR suggests that the Na$^+$ ion is “sandwiched” between two ammonia molecules at the centre of the octahedral interstice.

FIG. 3: $^2$H NMR spectra show typical Pake doublets at low temperatures both in the metallic and in the insulating sample. The gradual motional narrowing as the temperature rises, observed in the metallic sample, does not occur in the insulating one because of the hampered NH$_3$ rotation in it.

FIG. 4: Pictorial view of the (NH$_3$)$_2$NaK$_2$C$_60$ structure as inferred from NMR and x-ray measurements. For $x = 2$ the Na$^+$ ion is “sandwiched” between two ammonia molecules at the centre of the octahedral interstice.
assuming the same Na-N distance (2.5 Å), only a 1.5% reduction (from 2.29 to 2.26 Å) of the closest H-C distance takes place.

V. $^{13}$C NMR

The lack of a magnetic response in the insulating phase, as evidenced by SQUID measurements, suggests that this phase does not arise from a classical Mott transition as the one observed, for example, in NH$_3$K$_2$C$_60$. Whereas the latter follows a linear Korringa law, the insulator shows two thermally activated exponential relaxations, as reported in the inset. The fit with a single relaxation shown in the main figure (doted line) is characterized by an average $E_{\text{act}} = 71 \pm 5$ meV spin-gap.

![Figure 5: $^{13}$C NMR $1/T_1$ vs. temperature dependence in insulating and metallic (NH$_3$)$_2$NaK$_2$C$_60$. Whereas the latter follows a linear Korringa law, the insulator shows two thermally activated exponential relaxations, as reported in the inset. The fit with a single relaxation shown in the main figure (doted line) is characterized by an average $E_{\text{act}} = 71 \pm 5$ meV spin-gap.](image)

The presence of a singlet-triplet gap$^{20}$, like the one observed here, is typical of Jahn-Teller (JT) distorted C$_{60}^{-}$. This, together with the lack of magnetism and the presence of two different energy gaps, indicates that a quasi-static charge unbalance or, in chemical language, a disproportion as C$_3$+$^{2}$ → C$_2$$^{−}$ + C$_{60}^{-}$ could take place in our system. The different pre-exponential factors could account for two different superfine interactions of the paramagnetic electrons with the $^{13}$C nuclei. Their ratio ($A_1/A_2 \sim 5$) is close to the value (~4) reported for Na$_2$C$_{60}$ and K$_2$C$_{60}$.$^{20}$ The two activation energies, on the other hand, are quite close, in agreement with theoretical predictions.$^{23}$ It is interesting to note that recent studies by V. Brouet al., et also based on NMR relaxation measurements, report the observation of a similar JT induced spin gap not only in C$_{60}^{-}$ systems$^{23}$, but even in conducting Cs$_2$C$_{60}$ and superconducting Na$_2$CsC$_{60}$ and Rb$_2$CsC$_{60}^{24}$, in which the presence of C$_{60}^{-}$ anions with even charges is explained as the result of fast dynamical charge disproportions or fluctuations. If we suppose that the same effect could be present in the metallic phase of (NH$_3$)$_2$NaK$_2$C$_{60}$ (although it could not be observed in relaxation time measurements for other reasons$^{25}$), the known anomalies of this superconductor and the observed evolution to the insulating phase could be the effect of...
the slowing down, and eventually of a freezing, of these charge fluctuations.

The $^{13}\text{C}$ spectra taken at low temperature ($T = 20$ K), where narrowing due to molecular reorientational dynamics is ineffective, show in both cases Gaussian line-shapes. They are roughly centered at 200 ppm with respect to TMS (tetramethylsilane) and have different FWHM values: 118 ppm for $x \approx 0.9$ and 177 ppm for $x \approx 2.0$. Both the linewidths and their positions fully agree with previous observations: those for $x \approx 0.9$ doping conform to the values found in other superconducting fullerides$^{23}$, whereas the values for $x \approx 2$ doping agree with those measured in the $C_{60}$ and $C_{60}^\text{I}$ insulating systems$^{24,25}$. The linewidth observed in the metallic phase, smaller than that in the insulator, can be attributed to the different sign of the CSA with respect to the Knight shift anisotropy, yielding a line narrowing once the two tensors are added. These observations are also consistent with the insulating nature of the $x = 2.0$ phase.

VI. CONCLUSIONS

In conclusion we have shown that $(\text{NH}_3)_2\text{NaK}_x\text{C}_{60}$ for $x > 1$ evolves toward a rather singular insulating phase which, unlike other known cases, does not show any magnetic order. Moreover, this phase is hardly imputable to a lattice expansion but, more likely, it originates from a charge disproportion among $C_{60}$ anions, evidenced by the presence of a spin gap. According to theoretical analysis$^{26}$, in odd electron systems JT effect adds to the Coulomb repulsion to localize electrons in a non magnetic ground state, while the opposite happens in even electron systems. The related gain in energy could drive our system to favour a JT induced transition towards the insulating state rather than following the conventional Mott-Hubbard route. This system would represent the first example of a JT induced metal-to-insulator transition in superconducting fullerides.

Similar charge instabilities could also be at the origin of the anomalous $T_c$ vs. lattice parameter observed in the (super)conducting phase of our system. This suggestion is supported by the recent observation of a similar spin gap in the conducting CsC$_{60}$ and superconducting Na$_2$CsC$_{60}$ and Rb$_3$C$_{60}$, where a fast dynamical disproportion might be detectable already in the metallic phase.

Fundamental questions remain however open to deeper theoretical and experimental investigations, regarding the compatibility between the charge disproportion mechanism on one side and the non conventional BCS nature of the superconducting condensate on the other. This non conventional behaviour is clearly evidenced in the superconducting phase of the systems studied in the present work and confirmed also by recent experiments involving the density of states at the Fermi level or the critical magnetic fields and the field penetration depth$^{10}$.

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