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BIPOLARONIC MODEL AND CRITICAL FIELDS IN THE SUPERCONDUCTING STATE OF K_3C_{60}

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A model of "Fullerene" T_c's is proposed based on a strong electron-phonon interaction, which forms a charged Bose-liquid of small bipolarons. The experimental H_{c1} and H_{c2} data suggest that the bipolaronic picture may be relevant to describe M_xC_{60}. Some anomalous properties of these compounds are predicted.

In an interesting Letter [1] Holczer et al. presented measurements of temperature dependence of the lower H_{c1} and upper H_{c2} critical fields in superconducting K_3C_{60} and claimed that "experiments on the critical magnetic fields in K_3C_{60} show a strongly type-II superconducting state *with the temperature dependence of the parameters well described by mean-field theory*".

Contrary to the last remark, I intend to show that the temperature dependence of H_{c2} and H_{c1} measured by Holczer et al [1] clearly contradicts with the canonical mean-field BCS description and can be explained within the bipolaron theory of superconductivity [2].

The conclusion by Holczer et al [1] is based on a groundless assertion that "a linear dependence adequately fits the temperature dependence of H_{c2}". However just an unbiased observation of the experimental curves, Fig. 3,4 of Ref. [1] clearly reveals a nonlinear (upward) temperature dependence of H_{c2} in the temperature region comparable with that used in Ref. [1] for a linear fit. This upward temperature dependence can hardly be attributed to the sample inhomogenity or to the fluctuations because as pointed out in Ref. [1] "the magnetic transition is narrow, less than 1K, suggesting that the superconducting transition is relatively homogeneous".

My point is that due to its cluster structure K_3C_{60} is a system in which real-space electron pairs (small bipolarons) form like in a great variety of other compounds: Ti_4O_7, $Na_xV_2O_5$, WO_3, Chevrel phases and in high-T_c metal oxides. The ground state of carriers is a charged narrow band Bose-liquid [2] with the non-BCS temperature dependencies of H_{c2} [3] and H_{c1} [4]:

$$H_{c2} = H_2 \left(\frac{1 - t^{3/2}}{t} \right)^{3/2} \tag{1}$$

$$H_{c1} \simeq H_1(1 - t^2) \tag{2}$$

where $t = \frac{T}{T_c}$ and H_2, H_1 are temperature independent constants.

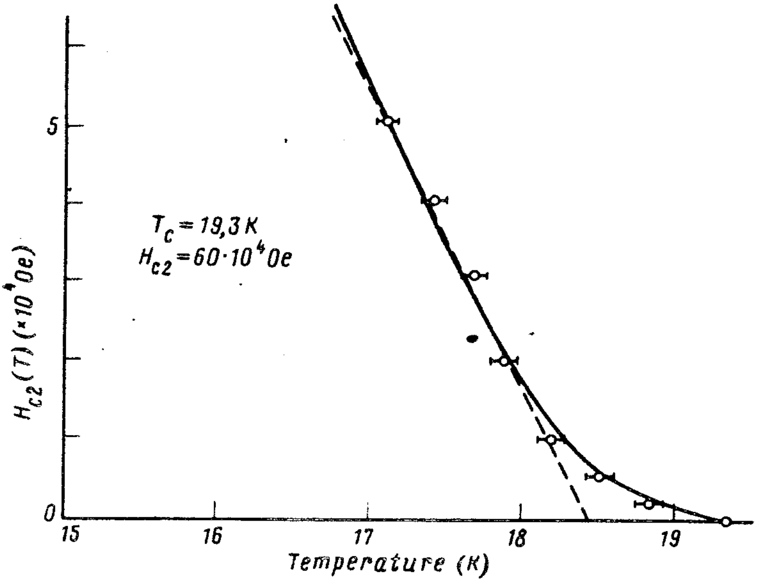

Fig.1. Temperature dependence of the upper critical field [1]. The solid line is a fit with eq. (1), the dashed line is a linear fit, used in Ref. [1]

As one can see from Fig.1 the non-mean-field temperature dependence of the upper critical field of a charged Bose gas scattered by impurities Eq.(1) fits the experiment [1] much better than the linear mean-field curve (dashed line).

As for H_{c1} it was already shown in Ref. [1] that the quadradic law, Eq.(2), describes the experimental data but not the two-fluid or BCS temperature dependence of H_{c1}.

Thus the quadratic temperature dependence of H_{c1} which was used in Ref. [1] as an empirical law, has a clear microscopic explanation as the temperature dependence of the lower magnetic field of small bipolarons.

Using the magnetic penetration depth for charged Bosons (charge 2e) $\lambda_H^2(0) = (8\pi ne^2/m^{**}c^2)$ [2,3] with the electron concentration $n = 5.7 \cdot 10^{21} 1/\mathrm{cm}^3$ and $\lambda_H(0) = 2400Å$ [1] one obtains the bipolaron mass $m^{**} = 24me$ which gives for the bipolaron bandwidth $D = z\hbar^2/a^2m^{**} \simeq 200\mathrm{K}$, with $z = 6$ being a coordination lattice number and $a \simeq 10Å$ being a lattice constant. Our formula for the critical temperature of the superconducting transition of small bipolarons (Eq.(3.26) of Ref. [3]) gives $T_c < D/6 \simeq 33\mathrm{K}$, which agrees rather well with the experimental values $T_c = 18\mathrm{K}$ (K_3C_{60}) [5] and $T_c = 30K$(Rb_3C_{60}) [6] .

In conclusion, the experimental data [1] does not support the claim of Ref. [1] mentioned at the beginning of this Letter and suggests that the bipolaronic picture is more relevant to describe the superconducting K_3C_{60}. The coherence length $\xi = 26Å$ estimated in Ref. [1], is also not large enough compared with $a = 10Å$ to apply the mean field theory (in Al, which is a classical BCS superconductor, $\xi/a = 10^4$). Of course, more experimental evidence is necessary before a definitive conclusion about the nature of carriers in K_3C_{60} can be made. Within the bipolaronic picture I predict: a λ-like heat capacity jump at T_c as in He^4, a strong pressure dependence of T_c, two gaps, the largest of which is temperature independent and exists *above* T_c.

I would like also to remark that the Coulomb repulsive interaction in M_xC_{60} may be strongly suppressed by a high value of the dielectric constant, as in metal oxides where the high-frequency ϵ is larger than 10. On the other hand the characterstic phonon frequency may be high ($\gtrsim$ 0.2eV). These factors make possible the existence of mobile bipolarons with a reasonable value of their effective mass contrary to Anderson's objection [7].

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