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ARGUMENTS FOR QUASI-ONE-DIMENSIONAL ROOM TEMPERATURE SUPERCONDUCTIVITY IN CARBON NANOTUBES

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ABSTRACT: In this article and references herein, I provide over twenty arguments for room temperature superconductivity in carbon nanotubes. The one-dimensionality of the nanotubes complicates the right-of-passage for prospective quasi-one-dimensional superconductors. The Meissner effect for individual tubes is less visible because the diameters of the tubes are much smaller than the penetration depth. Zero resistance is less obvious because of the quantum contact resistance and significant quantum phase slip, both of which are associated with a finite number of transverse conduction channels. Nonetheless, on-tube resistance at room temperature has been found to be indistinguishable from zero for many individual multi-walled nanotubes and a large Meissner effect has been observed in large bundles of multi-walled nanotubes at room temperature. On the basis of these arguments, I suggest that carbon nanotubes deserve to be classified as room temperature superconductors.

Key words: Room-temperature superconductivity, carbon nanotubes

1 Introduction

Finding room temperature (RT) superconductors is one of the most challenging problems in science. In 1999, Tsebro et al. [1] observed a substantial remnant magnetization in multi-walled carbon nanotubes (MWNTs) up to room temperature, which might be consistent with RT superconductivity if the authors could have been able to rule out ferromagnetic impurities in their samples. In November 2001, Zhao and Wang [2] provided subtle experimental evidence for RT superconductivity (SC) in MWNTs. Since then, Zhao [3, 4, 5, 6] has provided over twenty arguments for RTSC in both single-walled and multi-walled carbon nanotubes. Very recently, Kopelevich and coworkers [7] have given evidence for RTSC in graphite-sulfur composites.

It is generally believed that the superconducting transition temperature T_c cannot be higher than 30 K within the conventional phonon-mediated mechanism although there is no theoretical justification for the T_c limit. Alexandrov and Mott have demonstrated that Bose-Einstein condensation of bipolarons in polar materials can lead to a T_c higher than 100 K [8]. Ginzburg [9] and Little [10] have proposed that high-temperature superconductivity could be realized by exchanging high-energy electronic excitations such as excitons and plasmons. Lee and Mendoza have shown that superconductivity as high as 500 K can be reached through a pairing interaction mediated by undamped acoustic plasmon modes in a quasi-one-dimensional (1D) electronic system [11]. Moreover, high-temperature superconductivity can occur in a multi-layer electronic system due to an attraction of charge carriers in the same conducting layer via exchange of virtual plasmons in neighboring layers [12]. If these theoretical studies are relevant, one should be able to find high-temperature superconductivity in quasi-one-dimensional and/or multi-layer systems such as cuprates, carbon nanotubes and graphite-sulfur composites. In this article, I will review some arguments for RTSC especially in single-walled nanotubes (SWNTs). Over twenty detailed arguments for RTSC in both SWNTs and MWNTs, and alternative explanations to some data have been given in Refs. [3, 4, 5, 6].

2 Electrical transport and tunneling spectra in SWNTs

It is well known that, for quasi-1D systems disorder has extremely strong effects on electrical transport [13, 14]. For a noninteracting 1D system, all states get localized in the presence of an infinitesimal random potential [15]. Interactions can modify this picture, as shown clearly from theoretical works on one-chain and two-chain systems on the basis of bosonization and renormalization-group techniques [13, 14]. It is shown that metal-like conductivity occurs only for strongly attractive interactions that lead to quasi-1D s-wave high-temperature superconductivity [13, 14]. In contrast, for repulsive interactions d-wave superconductivity would occur for the pure system and, in the presence of disorder and/or impurities, the resistivity decreases monotonically with increasing temperature (semiconductor-like behavior) [14]. Thus, quasi-1D d-wave superconductors due to repulsive interactions behave like insulators at low temperatures [14]. Alternatively, the metal-like conductivity below a mean-field superconducting transition temperature T_{c0} in quasi-1D s-wave superconductors can be understood in terms of quantum phase slips (QPSs). Indeed, QPS theories [16, 17] can well explain a number of experiments that show a metal-like temperature dependence of the resistance well below T_{c0} in thin superconducting wires [16, 18, 19].

Within a QPS theory [17], the intrinsic on-wire (on-tube) resistance $R_i \propto T^{2\mu-3}$ for $\Phi_\circ I/ck_B << T < 0.5T_{c0}$, and $R_i \propto I^{2\mu-3}$ (independent of temperature) for $T << \Phi_\circ I/ck_B$. Here $\Phi_\circ$ is the quantum flux, c is the speed of light, I is the measuring current, and μ is a quantity that characterizes the ground state. The zero temperature resistance can approach zero when $\mu > 2$ and the current approaches zero, but is finite when $\mu \leq 2$. When μ <1.5, R_i increases with decreasing temperature (insulating behavior), while for $\mu > 1.5$, R_i decreases with decreasing temperature (metallic behavior). Only if the QPSs are strongly suppressed, can zero on-tube resistance be approached below T_{c0}.

Considering the contact resistance and the on-tube resistance due to QPSs, we obtain the two-probe resistance of a superconducting tube:

$$R(T) = R_0 + aT^p. \tag{1}$$

Here $p = 2\mu - 3$, $R_0 = R_Q/tN_{ch} + 2R_c$, t is the transmission coefficient ($t \leq 1$), $R_Q = h/2e^2$ =12.9 kΩ is the resistance quantum, and R_c is the contact resistance. The coefficient a normalized by the tube length L is given by [17]

$$a/L \propto \exp(-2S_{core}), \tag{2}$$

where

$$S_{core} \propto [N(E_F)]^{1/3}\sigma^{2/3}\Delta^{2/3}, \tag{3}$$

where $N(E_F)$, σ and Δ are the density of states at the Fermi level, the normal-state conductivity and the superconducting gap, respectively. It is apparent that the quantity a/L is very sensitive to S_{core} and thus to σ when S_{core} is significant. Thus, a/L should strongly depend on disorder of tubes.

Now we compare the QPS theory with the resistance data of SWNTs that are known to exhibit quasi-1D superconductivity. For the smallest diameter SWNTs with d = 0.42 nm, T_{c0} was found to be about 15 K from both magnetic and electrical measurements [20]. In Fig. 1a, we plot the two-probe resistance as a function of T/T_{c0} for this SWNT. It is apparent that the resistance increases more rapidly above $0.5T_{c0}$ and flattens out towards T_{c0}. Above T_{c0}, the resistance decreases with increasing temperature [20]. The broad transition and finite resistance at any finite temperatures below T_{c0} are caused by a large superconducting fluctuation in quasi-1D systems [20]. Zero on-tube resistance state is only possible at zero temperature [20, 17].

As demonstrated in Fig. 1b, the temperature dependence of the resistance below $0.5T_{c0}$ can be well fit by Eq. 1. From the fit, we find that p = 1.77±0.18. This suggests that the QPS theory can indeed explain the quasi-1D superconductivity in this SWNT. Using $p = 1.77$ and $p = 2\mu - 3$, we obtain $\mu = 2.4 > 2$, implying zero on-tube resistance at zero temperature [17].

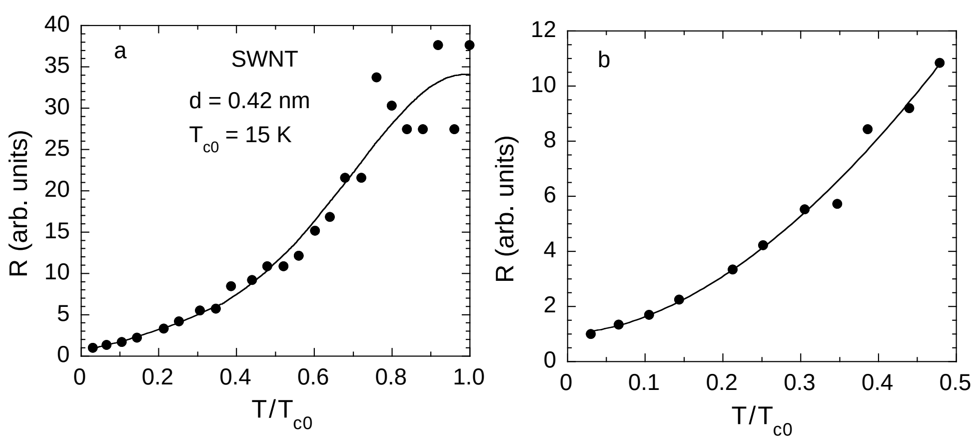

Figure 1: a) The resistance as a function of T/T_{c0} for the smallest diameter SWNTs with $d = 0.42$ nm. The data are extracted from Ref. [20]. b) The temperature dependence of the resistance below $0.5T_{c0}$.

In Fig. 2, we show the temperature dependences of the resistance at zero gate voltage for two metallic chirality SWNTs with different lengths (L= 200 and 4000 nm, respectively) and similar diameters (d = 1.5 and 1.7 nm, respectively). The contacts to the nanotubes are nearly ideal with the transmission probability close to 1 (Ref. [21, 22]). In this case, the two-probe resistance approaches 6.45 kΩ if the on-tube resistance approaches zero. It is remarkable that the temperature dependences of the resistance can be well fit by Eq. 1 with p = 1.71±0.23 for L = 200 nm and p = 1.48±0.11 for L= 4000nm. The exponents are very close to the one (p = 1.77±0.18) found for the smallest SWNTs with T_{c0} = 15 K. If we use the fixed parameter p = 1.48 for both L= 200 and 4000 nm tubes, the best fits show that the coefficient a/L for L = 200 nm tube is about 10 times larger than that for

L = 4000 nm. Such a large difference in the a/L values of the two tubes can be only explained by Eq. 2 and Eq. 3 that are predicted from QPS theory for quasi-1D s-wave superconductivity. This is because a small difference in the normal-state conductivity can lead to a huge difference in the a/L values according to Eq. 2 and Eq. 3. On the other hand, due to phonon backscattering and/or electron umklapp scattering in a Luttinger liquid (LL) with repulsive interactions, the on-tube resistivity is theoretically shown to exhibit a semiconductor-like behavior at low temperatures and a power-law behavior with $p \simeq 0.6$ at high temperatures [23, 24]. None of the theoretically predicted features is consistent with the data in Fig. 2. Further, the inelastic mean free path at room temperature for L = 200 nm tube would be about 160 nm if the tube were not a quasi-1D s-wave RT superconductor [6]. Such a short inelastic mean free path at RT for L= 200 nm tube and a huge difference in the a/L values of the two similar tubes are not consistent with any inelastic scattering processes in carbon nanotubes.

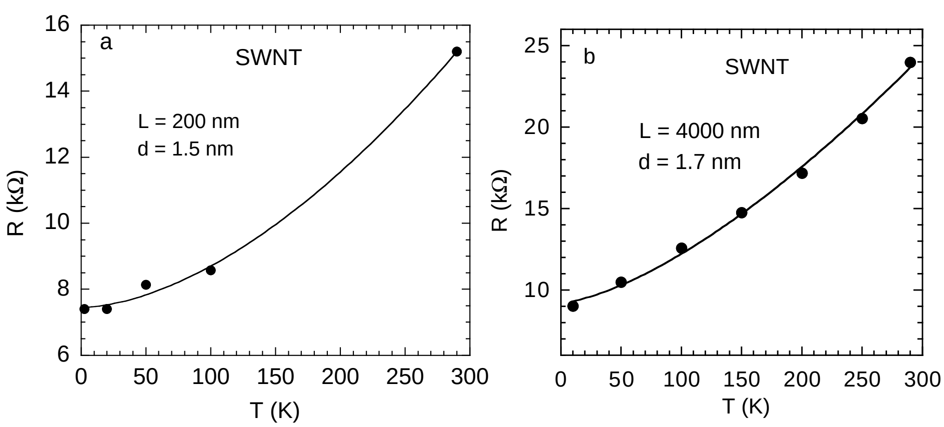

Figure 2: The temperature dependences of the resistance at zero gate voltage for two SWNTs with L= 200 and 4000 nm. The data are extracted from Refs. [21, 22].

For another tube with L = 800 nm, the resistance at zero gate voltage is independent of temperature below 270 K (Ref. [21]), i.e., p =$2\mu - 3 \simeq 0$. It was also shown that [21] underdoping leads to p <0 while overdoping gives rise to $p > 0$. This doping dependence of p is only consistent with the QPS theory. Within the QPS theory [17], $\mu \simeq 50r/\lambda_L \propto \sqrt{n}$ (where λ_L is the London penetration depth and n is the carrier density). It is clear that

μ increases with n such that p can cross over from a negative value at low doping to a positive one at high doping, in agreement with experiment [21].

Now we discuss tunneling spectra. From a single-particle tunneling spectrum obtained through two high-resistance contacts (see Fig. 1b of Ref. [25]), we can clearly see a pseudo-gap feature that appears at an energy of about 220 meV. Having ruled out the LL behavior (see the above discussion), we could relate the pseudo-gap feature to the superconducting gap smeared by QPSs. Considering the broadening of the gap feature due to QPSs and the double tunneling junctions in series, we estimate the superconducting gap $\Delta(0)$ to be about 100 meV. The scanning tunneling microscopy and spectroscopy [26] on individual single-walled nanotubes also show pseudo-gap features with $\Delta(0) > 100$ meV in doped metallic SWNTs. Using $k_B T_{c0} = \Delta(0)/1.76$, we find $T_{c0} \simeq 660$ K.

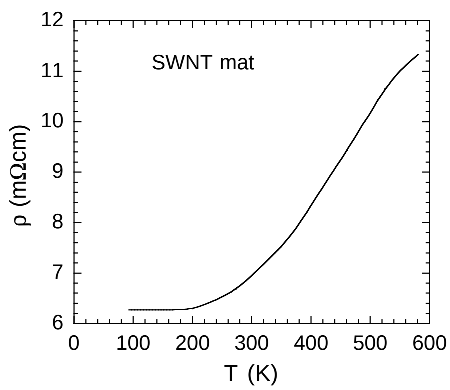

Figure 3: Temperature dependence of the resistivity for a SWNT mat with $T_{c0} \simeq 640$ K. The data are extracted from Ref. [27].

This gap value deduced from the tunneling spectrum is in good agreement with electrical transport data for a SWNT mat (see Fig. 3). Below 200 K the resistivity is nearly temperature independent while above 200 K the resistivity increases suddenly and starts to flatten out above 550 K. Such a temperature dependence is expected for granular superconductors [28] where superconducting islands are separated by non-superconducting materials and/or voids. For the SWNT mat, the nonsuperconducting materials are semiconducting chirality tubes. If the resistivity jump above 200 K were due to inelastic scattering, then the inelastic mean free path at 600 K would be less than 2 nm according to the resistivity difference between 600 K and 200 K. Such a short inelastic mean free path at 600 K is not compatible with any inelastic scattering processes in carbon nanotubes. Alternatively, by comparing Fig. 3 with Fig. 1a, we find that this temperature dependence of the resistance agrees with quasi-1D superconductivity with a $T_{c0} \simeq 640$ K.

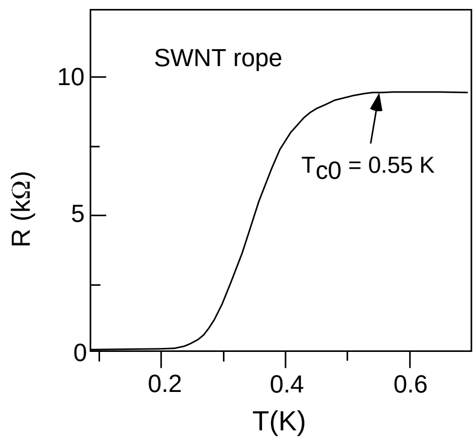

Figure 4: The temperature dependence of the resistance for a SWNT rope with $T_{c0} = 0.55$ K. The figure is reproduced from Ref. [29]

The electrical transport and single-particle tunneling spectra thus consistently suggest that T_{c0} in SWNTs can be higher than 500 K. Nevertheless, a much lower T_{c0} (∼0.55 K) was observed in a SWNT rope [29]. The rope consists of about 400 individual SWNTs with $d \simeq 1.5$ nm. Fig. 4 shows the resistance data for the SWNT rope. One can see that the resistance starts to drop below about 0.55 K. It is interesting to note that the resistivity of the rope decreases with increasing temperature up to 300 K in the normal state, and that the elastic mean free path just above T_{c0} is very short (8 nm) [29], similar to the case of the smallest SWNTs [20]. This suggests that these nanotubes behave like semiconductors in the normal state and like metals in the superconducting state. Very low superconductivity in this SWNT rope may be due to the fact that the tubes are very lightly doped. This is in agreement with the tunneling spectrum, which shows no pseudo-gap feature at 5 K in an undoped or very lightly doped armchair tube [30].

3 Raman scattering in a SWNT rope

It is known that Raman scattering has provided essential information about the electron-phonon coupling and the electronic pair excitation energy in the high-T_c cuprate superconductors [32, 33, 34]. The anomalous temperature-dependent broadening of the Raman active B_{1g}-like mode of 90 K superconductors $RBa_2Cu_3O_{7-y}$ (R is a rare-earth element) allows one to precisely determine the superconducting gap [33]. The pronounced softening observed only for the B_{1g} mode is due to the fact that the phonon energy of the B_{1g} mode is very close to $2\Delta(0)$ and the mode is strongly coupled to electrons [33, 35].

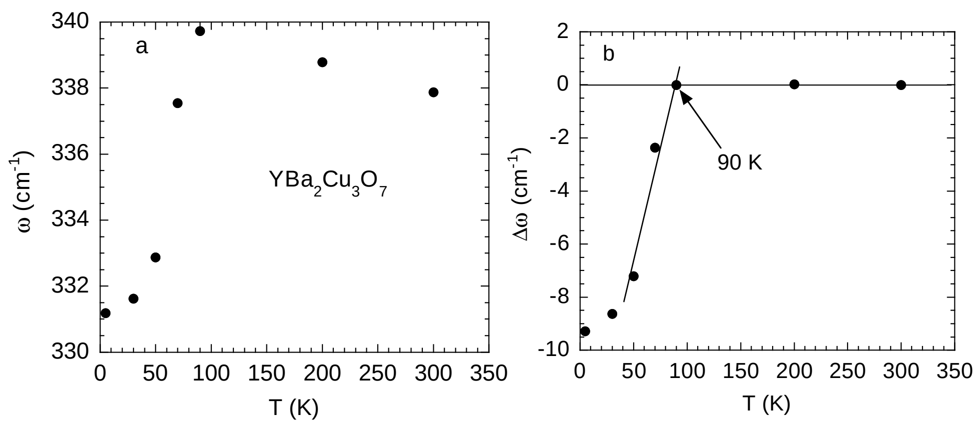

Figure 5: a) Temperature dependence of the frequency for the Raman-active B_{1g} mode of a 90 K superconductor $YBa_2Cu_3O_{7-y}$. The data are extracted from Ref.[32]. b) The difference between the measured frequency and the linearly fitted curve above T_c.

The temperature dependence of the frequency for the Raman-active B_{1g} mode of a 90 K superconductor $YBa_2Cu_3O_{7-y}$ is shown in Fig. 5a. It is apparent that the frequency decreases linearly with increasing temperature above T_c and that the mode starts to soften below about $0.95T_c$. The temperature dependence of the frequency above T_c is caused by thermal expansion. The temperature dependence of the frequency will become more pronounced at higher temperatures because the magnitude of the slope $-d\ln\omega/dT$ is essentially proportional to the lattice heat capacity that increases monotonically with temperature. The significant softening of the mode below T_c occurs only if the energy of the Raman mode is very close to $2\Delta(0)$ and the electron-phonon coupling is substantial [35], as it is the case in the 90 K superconductor $YBa_2Cu_3O_{7-y}$ [32, 33, 34]. In order to see more clearly the softening of the mode, we show in Fig. 5b the difference of the measured frequency and the linearly fitted curve above T_c. It is clear that the softening starts at about 89 K $\simeq 0.95T_c$.

Fig. 6a shows the temperature dependence of the frequency for the Raman active G-band of a SWNT rope. It is striking that the frequency data show a clear tendency of softening below about 630 K. Above 630 K, the

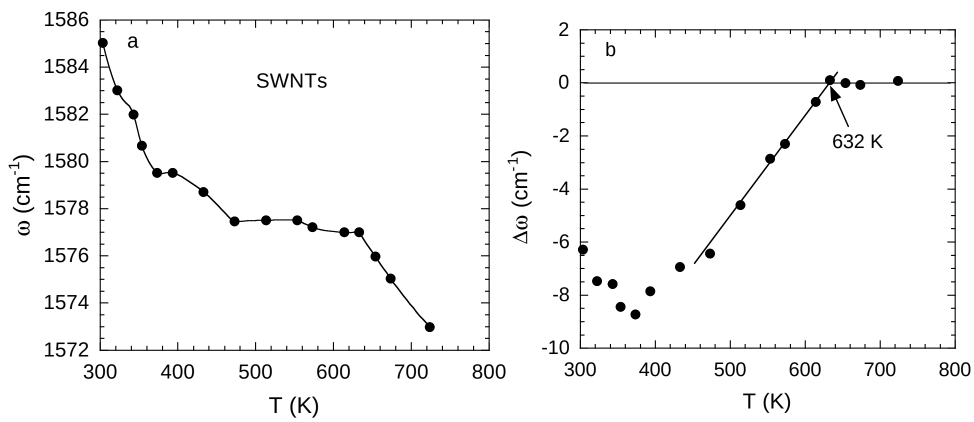

Figure 6: a) Temperature dependence of the frequency for the Raman active G-band of a SWNT rope. The data are from Ref. [31]. b) The difference of the measured frequency and the linearly fitted curve above the kink temperatures (see text).

frequency decreases linearly with increasing temperature with a slope much larger than that in $YBa_2Cu_3O_{7-y}$. This is due to a much larger thermal expansion at such high temperatures in SWNTs. The slope will decrease with decreasing temperature and eventually level off at low temperatures. The leveling off between 450-630 K must be due to the competition between the mode hardening due to the normal thermal effect and the mode softening possibly due to a superconducting transition or a transition to charge/spin density wave. The transition to charge/spin density wave will lead to a metal-insulator transition upon cooling, in contrast to the transport data in Fig. 3.

In order to see more clearly the softening of the mode, we show in Fig. 6b the difference between the measured frequency and the linearly fitted curve above the kink temperature (~630 K). One can see that the result shown in Fig. 6b is similar to that shown in Fig. 5b. This suggests that the softening of the Raman active G-band in the SWNTs may have the same microscopic origin as the softening of the Raman active B_{1g} mode in $YBa_2Cu_3O_{7-y}$. This explanation is plausible only if the phonon energy of the G-band is very close to $2\Delta(0)$. We are fortunate that the phonon energy of the G-band is 197 meV, very close to $2\Delta(0) \simeq 200$ meV deduced above from the tunneling spectrum and the electrical transport experiment. Therefore, it is very likely that the softening of the Raman mode in the SWNTs is related to a superconducting phase transition.

From Fig. 6, we can clearly see that the softening starts at about 632 K. Using the fact that the softening starts at $0.95T_{c0}$ (Ref. [35]), we can assign the mean-field transition temperature T_{c0} = 665 K. It is interesting to note that there is a clear minimum at T^* = 370 K = $0.57T_{c0}$ in Fig. 6b. It is remarkable that such a minimum is also seen at about $0.6T_{c0}$ in a calculated curve for a superconductor (see Fig. 8 of Ref. [35]). This shallow minimum in the frequency shift is related to a sharp minimum in the real part of the polarization $\Pi(\omega, T)$, which occurs at $\hbar\omega/2\Delta(T^*) = 1$ for weak coupling [35, 36]. From the temperature dependence of the BCS gap [37], we find that $\Delta(T^*) = \Delta(0.57T_{c0}) = 0.93\Delta(0)$. With $\hbar\omega/2\Delta(T^*) = 1$ and $\hbar\omega$ = 197 meV, we finally obtain $\Delta(0)$ = 105 meV, which is in excellent agreement with the value deduced from the tunneling spectrum [4]. Then we calculate $2\Delta(0)/k_BT_{c0} = 3.66$, which is in excellent agreement with the prediction based on the plasmon-mediated pairing mechanism [11]. Moreover, the electron-phonon coupling constant of the hardest optic mode deduced from the Raman data is in quantitative agreement with an independent theoretical prediction [6].

4 Meissner effect and Remnant Magnetization in MWNTs

It is well known that the diamagnetic susceptibility of graphite is very small when the magnetic field is along the plane. It is also shown [38] that the diamagnetic susceptibility is small ($\sim 10^{-6}$ emu/g) when the field is along the tube axis direction. This theoretically predicted value is about one order of magnitude smaller than the measured one (-1.1×10^{-5} emu/g) [39]. This large discrepancy can be naturally resolved if the MWNTs are superconductors that contribute to diamagnetism due to the Meissner effect.

For tubes of radius r with magnetic field parallel to the tube axis direction, the zero-temperature diamagnetic susceptibility due to the Meissner effect is given by

$$\chi_{\parallel}(0) = -\frac{\bar{r^2}}{32\pi\lambda_{\theta}^2(0)}. \tag{4}$$

Here r is the radius of tubes, $\bar{r^2}$ is the average value of r^2, and $\lambda_\theta(0)$ is the penetration depth when the screening current is along the circumferential direction. The above equation is valid only if $\lambda_\theta(0)$ is larger than the maximum radius of tubes. If we assume that the radii of tubes are equally distributed from 0 to 100 Å, we find $\bar{r^2}$= 6000 Å^2. With the weight density of 2.17

g/cm^3 [40] and $\chi_{\|}(0) = -0.93 \times 10^{-5}$ emu/g [39] (we have subtracted the normal-state diamagnetic susceptibility), we calculate $\lambda_\theta(0) \simeq 1724$ Å. This value of the penetration depth corresponds to $n/m^*_\theta = 0.96{\times}10^{21}/\mathrm{cm}^3 m_e$, where m^*_θ is the effective mass of carriers along the circumferential direction. With $n = 1.6{\times}10^{19}/\mathrm{cm}^3$ (Ref. [41]), we find that $m^*_\theta \simeq 0.017\ m_e$, which is larger than the in-plane effective mass of 0.012 m_e for graphites [42]. Since the effective mass along the tube axis direction is nearly zero [43], the value of m^*_θ indicates that MWNTs are quasi-1D superconductors.

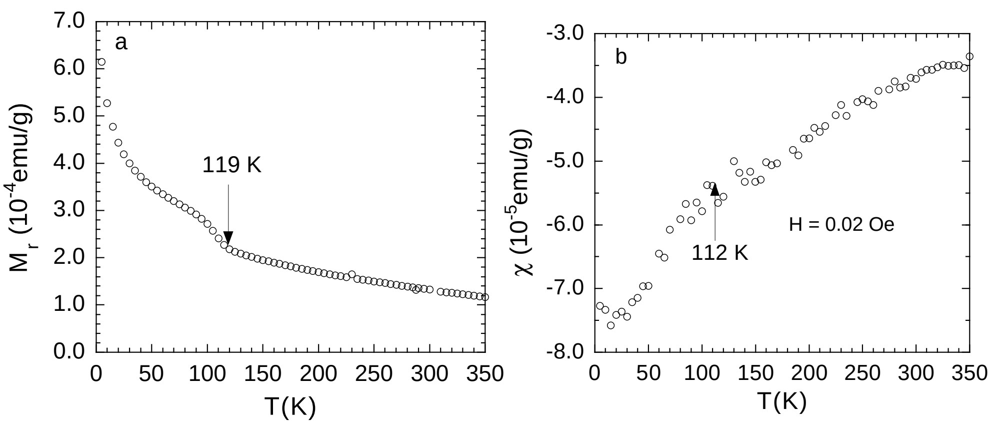

Figure 7: a) Temperature dependence of the remnant magnetization for multi-walled nanotube mat. b) The field-cooled susceptibility as a function of temperature in a field of 0.020 Oe. After Ref. [2].

To further show that MWNTs are room-temperature superconductors, we show in Fig. 7 the temperature dependencies of the remnant magnetization M_r and the diamagnetic susceptibility for our MWNT ropes. It is apparent that the temperature dependence of M_r (Fig. 7a) is similar to that of the diamagnetic susceptibility (Fig. 7b) except for the opposite signs. This behavior is expected for a superconductor. Although the M_r was also observed by Tsebro *et al.* up to 300 K [1], the observation of M_r alone does not give unambiguous evidence for RT superconductivity since such a M_r could be caused by ferromagnetic impurities.

We now rule out the existence of ferromagnetic impurities. If there were ferromagnetic impurities, the total susceptibility would tend to turn up below 120 K where the M_r increases suddenly. This is because paramagnetic susceptibility and M_r should increase simultaneously for ferromagnetic impurities. In contrast, the susceptibility suddenly turns down rather than turns up below 120 K (Fig. 7b). This provides strong evidence that the observed M_r in our MWNTs is not associated with the presence of random ferromagnetic impurities but with superconductivity. Moreover, the anomalies at about 120 K in the diamagnetic susceptibility and remnant magnetization correspond to the anomalies in both the conductance and Hall coefficient [3]. All these results can be consistently explained by RTSC [3].

4 Concluding remarks

Although the present article and Refs. [2, 3, 4, 5, 6] have provided over twenty arguments for RTSC in carbon nanotubes, and we cannot find any published data that are contradictory with the RTSC explanation, the ultimate proof of RTSC in carbon nanotubes should be the observations of a large Meissner effect and zero resistance at RT. As discussed above, the Meissner effect for isolated tubes is intrinsically small due to the fact that the penetration depth is much larger than the tube diameters. For Josephson-coupled large bundles of superconducting MWNTs, there should be a substantial Meissner effect. Indeed, I was told in this ARW conference that a Russian group has observed a large Meissner effect at RT in large bundles of MWNTs. Further, a negligible on-tube resistance ($<$86 Ω/μm) has been observed at RT in the majority of individual MWNTs [44, 45]. Another independent group [46] has also observed nearly zero on-tube resistance over a 4 μm long MWNT at RT ($<$1.0 Ω/μm). These results cannot be explained by ballistic transport at RT [6], but are consistent with quasi-1D RTSC.

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