Detection of a coherent boson current in the normal state of a high-temperature superconductor $YBa_2Cu_3O_y$ film patterned to micrometer-sized rings

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(Received 29 October 1997; revised manuscript received 25 March 1998)

The key to clarifying the mechanism of the appearance of high T_c in the high-temperature superconductor seems to be related to the anomalous properties in its normal state [H. Yasuoka, S. Kambe, Y. Itoh, and T. Machi, Physica B **199-200**, 278 (1994); H. Ding T. Yokoya, J. C. Campuzano, T. Takahashi, M. Randeria, M. R. Norman, T. Mochiki, K. Kadowaki, and J. Giapintzakis, Nature (London) **382**, 51 (1996); and T. Ito, K. Takenaka, and S. Uchida, Phys. Rev. Lett. **70**, 3995 (1993)]. Related to this is the presence of Bose-electron-type species such as paired holes [V. J. Emery and S. A. Kivelson, Nature (London) **374**, 434 (1995)] and holons [P. W. Anderson, G. Bakaran, Z. Zou, and T. Hsu, Phys. Rev. Lett. **58**, 2790 (1987)] in the normal state which has been proposed by theoreticians. We have succeeded in detecting the coherent Bose species in the normal state of $YBa_2Cu_3O_y$. The idea [L. P. Levy, G. Dolan, J. Dunsmuir, and H. Bouchiat, Phys. Rev. Lett. **64**, 2074 (1990)] of having the detected oscillation of a fermion persisting current caused by the Aharanov-Bohm effect on magnetic field scanning in the assemblage of the mesoscopic rings of metal was employed in the assemblage of a ring patterned in the $YB_2Cu_3O_y$ film. The sensitivity of our detection system is not high enough to detect fermions, but it is high enough for detecting coherent bosons if they exist. Oscillations with flux periods corresponding to $h/2e$ have been found at $T>Tc$, even at temperatures 30 K or more above T_c inserted in the patterned $YBa_2Cu_3O_y$ film, but have not been found in the patterned Au or Pd films. To explain the coherent current circulating in a ring with a circumference as long as 40 μm, presumably in the prestage to the superconducting stage, the possible coexistence of a minority of coherent bosons and a majority of incoherent bosons is discussed. The detection of minority coherent bosons in the background of a majority of incoherent species is possible in this method. [S0163-1829(98)08129-6]

The problem of why and how high-temperature superconductivity manifests itself has not been resolved yet after ten years since its discovery, and is still the hot problem in solid state physics. One of the recent interests of researchers seems to be the anomalous properties of the normal phase of the high-temperature superconductor (HTSC) observed by NMR,[1] x-ray photoemission spectroscopy (XPS),[2] and conductivity[3] [recently also in Raman scattering, see G. Blumberg *et al.*, Science **278**, 1427 (1997)] which are considered to be the cause of the anomolously high T_c. Based on these experimental results, Emery and Kievelson[4] discussed the striking possibility of the presence of a paired hole above T_c, that is, in HTSC's the pairing occurs at T_{MF} higher than T_c, in contrast to $T_{MF}=T_c$ in the conventional superconductor. The one paired hole with spin 0 is the same as one Bose particle or boson. Another type of boson probable in $T>T_c$ was proposed by Anderson[5] just after the discovery of HTSC. In the resonating-valence-bond (RVB) model, holes are separated into holons, charge carriers with spin 0, and spinons, spin carriers with no charge. This holon may be a boson. These kinds of bosons at $T>T_c$ have been proposed theoretically, but could not be confirmed experimentally. We believe that we have succeeded in directly detecting the bosons in HTSC's at $T>T_c$.

The idea we employed in detecting the boson current is the same as the one that Levy *et al.*[6] used in detecting persisting fermion current. In a ring in which the mean free path of the electron between the two inelastic scattering events is longer than the circumference, persistent current exists and oscillates according to the Aharonov-Bohm effect on magnetic field scanning with a period of $2\pi\phi/\phi_0$, where ϕ is the flux threading the hole of the ring and ϕ_0 the magnetic flux quantum. Levy *et al.*[6] observed this oscillation in an array of 1×10^7 mesoscopic isolated copper rings at a few tens mK. Technically, detecting such fermion current is very difficult because only one electron is effectively detected in one ring.[7] In contrast to this, many boson carriers can occupy the same energy level at the same time, reducing this difficulty much, so long as the phase coherence among the bosons is established. We have measured the magnetic moment of the array

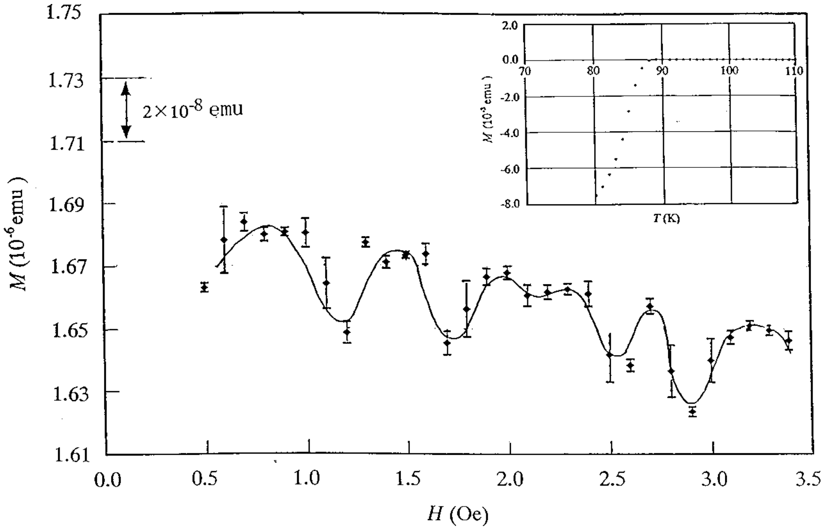

FIG. 1. Magnetization vs magnetic field at 95 K in a 0.3 μm thick YBCO film of $T_c = 88$ K. The films are patterned to the assemblage of microscopic rings as written in the text. This sample showed a twin pattern similar to that in single crystal Y123 by polarized optical microscope observation. The value at one magnetic field is an average of five-times repeated measurements ($n = 5$; n in the text). The oscillation on magnetic field scanning is clearly seen. The solid line curve is only a guide to the eye. The inset is the measured diamagnetism of this patterned film, showing $T_c = 88$ K.

of the isolated microsize ring of $YBa_2Cu_3O_y$ (Y123) in the normal state using a superconducting quantum interference device (SQUID) and succeeded in the detection of the oscillation, presumably due to the coherent bosons.

Before proceeding to the description of the experimental procedure, the magnetization in our system is estimated if the only e in one ring is effectively detected as in the case of a fermion. The current circulating ring I is eVf/L, where V is the velocity of the carrier e, f is the factor that the carrier's real path is extended by elastic scattering, and L is the length of the circumference of the ring. The magnetization M of the system is $M = NIS$, where N is the number of the ring and S is the area of the hole of the ring in which the flux is threaded. An example of an experimental parameter is $N = 1\times10^5$, $L = 40$ μm, and $S = 1\times10^{-6}$ cm^2. If we set $V = 5\times10^6$ cm/sec, which corresponds to the velocity for the Bose electron at 100 K ($kT = 0.01$ eV), and $f = 1$, M is calculated to be 2×10^{-12} emu. ($M = 4\times10^{-11}$ if we use the speed at the Fermi surface.) However, our lowest limit of the detection system for M is about 1×10^{-8} emu. So, detecting one fermion using our detection system is impossible. We need a coherent charge of 1×10^4 at least.

The dimensions of one unit of the rings are a ring width of 3 μm, an outer and inner circumference of 48 and 24 μm, respectively, and a hole dimension of 6×6 μm^2. The rings are arrayed with a center-to-center distance of 20 μm. The thin film of 0.3 μm thick Y123 was deposited on a MgO single crystal substrate of 0.5 mm thickness. The ring pattern was drawn using the photolithography method on the resist coating the Y123 film, and printed on the film later by ion-bombardment etching. Measurement of the magnetization was made using a SQUID XE-II of Quantum Design Co. The residual magnetic field was reduced to lower than 10 mG by a flux-gate meter before the measurement. The EDC system gave small fields with 0.1 G step by using a coil wound inside the superconducting (SC) magnet coil. (Sometimes a fixed field was set by a SC coil in addition to EDC to get a good SQUID response.) The data were taken in RSO mode. For each point of the magnetic field the measurements were made 5–10 times. A typical example of one unit of the measurement is the average of four 19 cycle RSO oscillations in 1.5 Hz and 3 cm amplitude. The sequential repetition of this unit by n (usually 5–10) times measurements was made for one magnetic field point in the field scanning of the 0.1 Oe step. The averages of this n-times measurement x_0 and the average deviation $\Sigma|x_n - x_0|/n$ are plotted in the figures of the present paper. The usual specimens were of the size 5×5 mm^2, with two to four pieces layered to the total thickness below 2 mm.

In Fig. 1, the result for a YBCO sample by using five times repeated measurements at one magnetic point ($n = 5$, n mentioned above) is shown. As shown in the inset, the T_c of this patterned film measured by diamagnetism is 88 K. The nearly sinusoidal oscillations are clearly seen in Fig. 1. The flux period of this oscillation δB calculated from $\delta BS = \phi_0$ is 0.58 G, where S is the area of the hole of the ring 6×6 μm^2 and ϕ_0 is magnetic flux quantum $ch/2e$, 2.07×10^{-7} G cm^2. If the effective S is used as 7×7 μm^2 the flux period is 0.42 G. The experimental results in Fig. 1 show that the δB's are 0.4 to 0.5 G. In another sample the flux periods were 0.5 G and did not exceed 0.6 G in any samples. This variation of the flux period seems to depend on the effective area of the ring hole, which mostly depends on the patterning process. It can be said that the observed flux periods agree well with the calculated value, which will lead immediately to a conclusion that the species we are observing at $T > T_c$ has $2e$, the same as that in $T < T_c$. The oscillation amplitude seen in Fig. 1 is not always uniform with the flux. This seems to be partly due to the fact that the area of the current path in the ring is too large compared to that of the ring hole. The size of the ring was determined by reconciliating the need to make the ring as small as possible and to reduce the technical difficulty on the small size patterning. If the ratio of the area of the ring hole to that of the ring were to become reasonably improved, a uniform oscillation amplitude would be observed. The oscillation amplitude was found to depend on the quality of the Y123 film. The sample in Fig. 1 showed twin pattern[8] domains (before the ring patterning) under the polarization microscope which are generally observed in the single crystal of the orthorhombic phase of Y123. In this sample the widths of the domains were wider than 20 μm which are sufficient to include one ring in one domain. This means that for one ring, the film quality is as good as the twinless single crystal.

In the samples in which the twin patterns were not observed, a similar oscillation of 0.4 to 0.5 G flux periods were

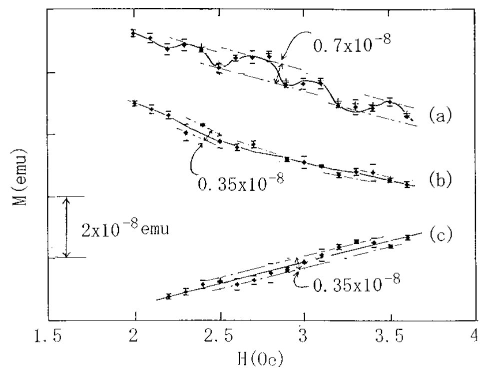

FIG. 2. Magnetization vs magnetic field is measured in a 0.3 μ thick ring-patterned film of $T_c = 78$ K at 110 K, shown in (a); in the patterned Au films of 500 Å thickness at 110 K in (b); and in a Pd film (not patterned) at room temperature in (c). The sample Y123 (before the ring patterning) did not show the twin pattern by optical microscope observation. A fixed field of 30 G was set by an SC coil to get a good SQUID response. All the data were taken by the repeated measurement of $n = 10$ for the one magnetic field. The averages and deviations are shown after peeling off the smallest and the largest points three times from $n = 10$, reducing to $n = 4$. However, the averages in the $n = 4$ do not change much from the averages of $n = 10$. The averages for $n = 10$ are plotted as plus signs only on the points where these symbols are not concealed by the average symbols for $n = 4$. The absolute values of M at 2.5 Oe are 1.24×10^{-6}, 1.05×10^{-6}, and 0.7×10^{-6} for (a), (b), and (c), respectively. The solid line curves are drawn only for a guide to the eye.

also observed, although the oscillation amplitude became much smaller. In Fig. 2, the case for a Y123 sample of $T_c = 78$ K measured at 110 K is shown in (a), in comparison with patterned-Au films measured at 110 K (b) and with a Pd film (not patterned) measured at room temperature (c). All the data in these cases were taken by the repeated measurement of $n = 10$ for one point of the magnetic field. The data in Fig. 2 are shown after peeling off the smallest and largest points three times from $n = 10$, which reduces n to 4. However, the averages in $n = 4$ do not change from the averages of $n = 10$. In this figure, the averages for $n = 10$ are plotted as plus signs only on the points where these average symbols are not concealed by the average symbol (♦) for $n = 4$. One may think that even in (c), and rather ambiguously in (b), small fluctuations may exist, but the maximum amplitudes of the fluctuations do not exceed 0.35×10^{-8} emu, in contrast to 0.7×10^{-8} emu in (a), and the fluctuation periods are not the same as those in (a). Such small fluctuations in (b) and (c) seem to be intrinsic in the SQUID measuring system in the range close to the detection limit. The oscillation signals clearly periodical with a period of about 0.4 G as shown in (a) were always observed in the ring-patterned Y123 samples, especially if the samples were kept for a long time, such as 15 h, at 110 K. It can be concluded that this oscillation at 110 K in the Y123 samples of $T_c = 78$ K is the same kind as that observed at 95 K in the ring-patterned sample of $T_c = 88$ K prepared from the twin-patterned film, although the amplitude of the former is much smaller than that of the latter, by about 3×10^{-8} emu.

In Fig. 3, a similar periodical oscillation in the extended range of 0–4 G is shown for $n = 5$ measurements at 110 K for another sample of $T_c = 78$ K. In this case data are reduced to $n = 3$ by the same procedure written above in the explanation of Fig. 2. As in Fig. 2, the averages for $n = 5$ are plotted as plus signs at the points where these symbols are not concealed by the average symbols for $n = 3$.

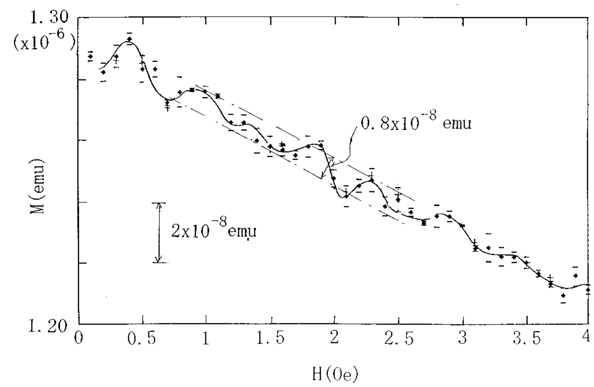

FIG. 3. Magnetization vs magnetic field is measured in a 0.3 μm thick Y123 ring-patterned film (the twin pattern was not observed) at 110 K. The sample was kept at 110 K for 17 h before the measurements. The measurements were repeated $n = 5$ times at each magnetic field. Data are reduced to $n = 3$ by the processing as stated in Fig. 2 and the text. As in Fig. 2, the averages for $n = 5$ are plotted as plus signs at the points where these symbols are not concealed by the average symbols for $n = 3$. The solid line curve is drawn only for a guide to the eye.

Summarizing the descriptions stated above in Figs. 1–3, it can be concluded that periodical oscillations with 0.4 to 0.5 G are observed only in the ring-patterned HTSC Y123, and even at temperatures 30 K higher than T_c. It is to be noted that this kind of oscillation, though smaller in amplitude, was recognized even at 120 K or above in some underdoped samples. We found a similar oscillation in Bi2212 at temperatures of 140 K.[9] In Ref. 2, Ding *et al.* reported that a pseudogap observed by ARPES in the underdoped samples exists at $T > T_c$ up to 170 K. Some researchers think the existence of such a pseudogap is related to the existence of the paired hole. Our results on these oscillations indicate the same tendency as the oscillation of this pseudogap.

The oscillation signals observed here must be due to the Bose particle current for the reason that we stated in the beginning of this paper. The fact that the coherent signal is observed may suggest that the inelastic scattering length must be larger than the circumference of the ring, 40 μm. But this length is much longer than those which are familiar in HTSC's. The scattering length l_0 is given by $l_0 = v\tau$, $\tau = h/(2\pi\lambda KT)$ (h: Planck's constant, λ: constant relating to the strength of the coupling between the phonons and the carrier, k: Boltzmann's constant). The largest λ is estimated to be 0.3,[10] which gives a l_0 of less than 100 Å. Some researchers think $\lambda \ll 0.3$. But is it possible to extend l_0 to 40 μm? We discuss this discrepancy in the following. In Table I, we give (1) the magnetization M if only one e with velocity v per ring exists, (2) the number of the boson carrier per ring to get $M = 1\times10^{-8}$ emu, (3) the number of the doped hole calculated from the well-accepted value of the density, $1\times10^{21}/\mathrm{cm}^3$, and (4) the De-Broglie wavelength, for three kinds of velocity (or energy), (a) 5×10^6 cm/sec, (b) 5×10^5 cm/sec, and (c) 5×10^4 cm/sec. The velocity (a) is the maximum velocity of Bose electrons at 100 K, calculated from $E = kT = 0.01$ eV. The velocity (b) just coincides with that of the sonic velocity in Y123.[11] Generally, a particle

TABLE I. This table gives the typical parameters of the assumed boson carriers to show that the existence of an experimentally detectable amount of coherent carriers, circulating a long path with a circumference of 40 μm, is reasonable.

	(a)	(b)	(c)
Velocity (v, cm/s)	5×10^6	5×10^5	5×10^4
and energy (eV) of e	1×10^{-2}	1×10^{-4}	1×10^{-6}
(1) M (emu) by one	2×10^{-12}	2×10^{-13}	2×10^{-14}
e per ring with v ($f=1$)			
(2) Numbers of e per ring	5×10^3	5×10^4	5×10^5
necessary for $M=1\times10^{-8}$ emu			
(3) Numbers of hole per ring		4×10^{10}	
(4) Wavelength of e (Å)	73	730	7300

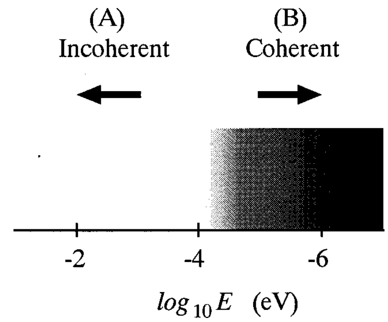

FIG. 4. This sketch shows an example of a scheme discussed in the text on boson carriers. The boson carriers are divided into incoherent and coherent parts with a boundary at 1×10^{-2} eV. Going to the right, the integrated phase of the collective boson carriers contained in one wavelength become more stable against the collision by incoherent bosons.

with charge e traversing a medium with a velocity slower than the sonic velocity cannot receive or give energy from or to the lattice, that is, the lattice phonons are not responsible for the inelastic scattering. Therefore, one condition to establish the coherence in our system is that the velocity is slower than (b) in Table I. Another condition is that the distance between the two carriers circulating the ring should be shorter than the wavelength of the carriers. The latter condition is also satisfied in (b) and (c). In Fig. 4, we give this scheme by a sketch that the carriers are divided to incoherent region (A) and coherent region (B) with the boundary at $E=1\times10^{-4}$ eV. In thermal equilibrium where the boson charge carriers react with lattice phonons effectively, the boson obeys the Boltzmann distribution. Therefore, around 100 K, the bosons are considered to be equally distributed in every energy. If so, the ratio of the number of bosons in region (B) is $1\times10^{-4}/0.01=1\%$ of the incoherent bosons in the region (A), that is, about 4×10^8 per ring. In region (B) where the energy transfer by a lattice phonon is impossible, the energy transfer by collision processes must weigh (B1) the collision between two bosons in the region (B), (B2) the collision with incoherent bosons in region (A), and (B3) collisions with spins.[3] The collision (B2) is indirectly related to lattice phonons, therefore the energy distribution may still be of Boltzmann type, that is, distributed equally in every energy. In region (B) the boson with the highest energy [Table I(b)] has a wavelength of 730 Å. The number of bosons of 4×10^8 is distributed in the circumference of 40 μm. If we consider that all particles in region (B) are centered at this energy, 550 bosons are included within one wavelength. In this one wave, one cannot distinguish the position of each boson. The phase integrated from the individual bosons will grow. This assemblage of bosons reacts as a whole to the single collision made randomly in time sequence by the bosons in region (A), and the energy of the integrated phase does not change much, as in the one-to-one incoherent boson collision. In other words, the phase becomes more stable for the collision. This effect makes it look as if the mean free path between the inelastic scatterings becomes longer. This phenomenon becomes enhanced at lower energy where the wavelength becomes longer. For instance, at an energy of 1×10^{-6} eV [Table I(c)] 55 bosons are included in one wavelength even if we limit the energy width to 1×10^{-6} eV. Such collective ordering seems to be similar to that at $T<T_c$ where all bosons are condensed into the lowest level. This state can be called the prestage of superconductivity. Thus, it is quite reasonable to assert that coherence bosons can be detected in the ring with a circumference as long as 40 μm. This conclusion can be made quite reasonably if we once postulate that the charge carriers in the normal state are bosons.

Thus, in the normal phase a majority of incoherent species and a minority of coherent species coexist as in the sketch of Fig. 4. The present method detects only the coherent current exclusively.

We have succeeded in the detection of boson current in the normal state of Y123 films patterned to micron-sized rings. The method of using the assembly of microscopic rings makes it possible to select minor coherent carriers in the background of the major incoherent carriers. In addition, an important merit of this method is that it can determine whether the carrier is of e or $2e$. For these reasons this method is unique in comparison to other conventional methods, i.e., resistance or optical measurements.

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