Search for a breaking of time-reversal symmetry in high-T_c superconductors

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A method is proposed for determining the internal magnetic field which would be produced by anyons in high-T_c superconductors. The idea is to measure the amplitude of even harmonics of the magnetic field which are generated by a nonlinear medium consisting of Bi 2212 single crystals with intergrowths of the 2223 phase. The experiments show that there is no average internal field in these samples, within 1 mG, at $T = 77$ K.

One of the intriguing new ideas which have been offered to explain high-T_c superconductivity is that under certain conditions these compounds contain particles with a fractional statistics, called "anyons."[1,2] The ground state of such particles would correspond to a breaking of spatial parity (P) and time reversal (T). One could thus observe a rotation of the polarization plane of light, a spontaneous Hall effect, and other effects which would stem from a breaking of T symmetry in these materials (Ref. 3, for example).

In the literature we find contradictory experimental results regarding the observation of magnetooptic effects in the high-T_c superconductors. Lyons *et al.*[4] have reported measurements of the circular dichroism upon the reflection of light from various single-crystal and film samples of high-T_c superconductors at $T > T_c$. Webber *et al.*[5]

measured the optical transparency of high-quality $YBa_2Cu_3O_{7-\delta}$ samples and the transmission of $Bi_2Sr_2CaCu_2O_8$ single crystals. They observed a circular optical effect at temperatures above the superconducting transition temperature T_c but below a characteristic temperature $T_s > T_c$. They interpreted their results as proof of the existence of states with a broken T symmetry below T_s. On the other hand, Spielman *et al.*[6] and Krichevtsov *et al.*[7] found no trace of a circular dichroism in $YBa_2Cu_6O_{7-\delta}$.

In this letter we are reporting a search for a state with a broken T invariance by a different method, which was not discussed in Ref. 3: a study of the generation of even harmonics of the magnetic field in single crystals of these superconductors.

1. If a sample has a broken T symmetry, and if there is not static magnetic field in the sample, the application of an external magnetic field h_β of frequency ω will result in the excitation of oscillations of the magnetization M_α at even harmonics:

$$M_\alpha = \chi^{(2)}_{\alpha\beta\gamma} h^\omega_\beta h^\omega_\gamma + \chi^{(4)}_{\alpha\beta\gamma\delta\lambda} h^\omega_\beta h^\omega_\gamma h^\omega_\delta h^\omega_\lambda. \quad (1)$$

There are several effects which might simulate the breaking of internal T parity and give rise to even harmonics: (1) a residual external magnetic field or pinned magnetic flux in the superconductor; (2) the finite frequency or dissipative processes in the crystals. We would thus like to detect the even harmonics by a method insensitive to effects of the second type. Effects of the first type, on the other hand, could be suppressed by canceling the external field. One such method is to study the nonlinear properties of single-crystal Bi(2-2-1-2) samples with intergrown layers of a Bi(2-2-2-3) superconductor. Such a structure constitutes a system of S–S'–S junctions, whose nonlinear susceptibility at the doubled frequency is proportional to the "internal" static field of the anyons. A system of this sort would make it possible to observe the "internal" field of the anyons not only in the case in which they have a ferromagnetic order but also in the case in which there is an antiferromagnetic field in neighboring planes. The reason is that the planes bordering the Josephson junction in the two different superconductors would create "internal" fields which are equal in magnitude but opposite in direction.

Measurements were carried out at $T = 77$ K on an apparatus for which the schematic diagram would be the same as that described in Ref. 8. Some additional features of the apparatus used in the present study were that (1) the cancellation of the first harmonic of the output signal was improved to a level of 60 dB, and (2) the measurement unit was placed at the center of a system consisting of three orthogonal pairs of Helmholtz coils. These coils made it possible to cancel the laboratory field to within

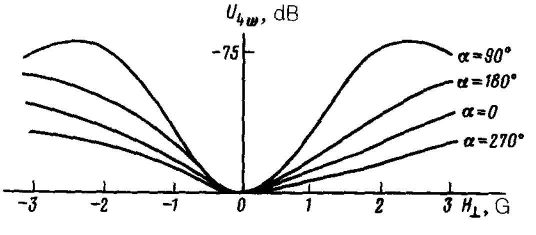

FIG. 1. Amplitude of the fourth harmonic versus the perpendicular field $H_\perp$ for a set of single-crystal samples.

$\pm$ 30 mG in the directions perpendicular to h_ω and to within $\pm$ 1 mG in the direction parallel to h_ω. As the condition for "complete" cancellation of the perpendicular components of the laboratory field we adopted the absence of an asymmetry from the $U_{2h\omega}(H)$ curve when the sign of the external field was reversed (Fig. 1).

The measurements were carried out by the following procedure. First, we canceled the laboratory field and inserted the test sample into the measurement coil. We then cooled the sample to 77 K. Measurements of the temperature dependence were carried out by evaporating liquid nitrogen from the system while simultaneously detecting the temperature and the amplitude of the measured signal on an x, y plotter.

The final cancellation of the external field was carried out on the basis of the amplitude of the second harmonic, with the help of a cylindrical sample of ceramic $Y_1Ba_2Cu_3O_7$. Consequently, the field was zero in the sample after the cancellation, but it would not be zero in free space. Because of the random orientation of the axes of the crystallites in a ceramic sample, all the internal magnetic fields should be averaged out. Our method for observing internal fields is a comparison method! Consequently, the assertion that there are no even harmonics in a zero external field and thus the assertion that there is no T breaking in single-crystal samples are valid to the extent that this assumption is accurate. When the test sample is placed in the measurement coil, a systematic "difference of zeros" may arise, because of (for example) differences among the demagnetizing factors of the ceramic sample, the field pickup, and the test sample. Because of these systematic differences, the field values corresponding to a zero amplitude of the second harmonic for the ceramic sample and for the single-crystal test sample might be different. This difference did not exceed 20 mG in the present experiments.

In single-phase Bi(2-2-1-2) single crystals with dimensions of $3\times3\times0.3$ mm, we found no harmonics in the spectrum of the output signal at frequencies over the interval 0.1–2 kHz.

In the layered structures, on the other hand, the spectrum of harmonics in the output signal was as rich as that in the ceramic, in which the nonlinearities are determined by internal Josephson junctions. The intensity of the even harmonics, $U_{2n\omega}$, depends on both the amplitude of the alternating field, h_ω, and the amplitude of the static external magnetic field, H. In particular, the presence of a field $H_\perp$ perpendicular to the c axis causes U_{2n} to depend strongly on the orientation of the crystal. It is thus especially important to cancel $H_\perp$ in order to observe effects of an anyon field. This cancellation was carried out until the dependence of U_{2n} became symmetric under reversal of the magnetic field. Figure 2 shows $U_{4\omega}$ vs $H_\parallel$ at $H_\perp = 0$ and 2.4 G for the same single-crystal sample as in Fig. 1.

To observe the internal magnetic field we used samples consisting of a stack of single-crystal Bi(2-2-1-2) + Bi(2-2-3-3) samples (Fig. 2). As we mentioned earlier, the 20-mG shift in the zero of the amplitude $U_{4\omega}$ upon a change in magnetic field ΔH is a methodological effect: A rotation of the sample through 180° changed neither the sign of ΔH nor its magnitude. It can be seen from these results that there are no grounds for asserting that there is an internal microscopic field stronger than 10^{-3} G in these single-crystal Bi(2-2-1-2) samples. If a field did indeed exist, and if it were directed oppositely in different parts of the samples (an antiferromagnetism), then

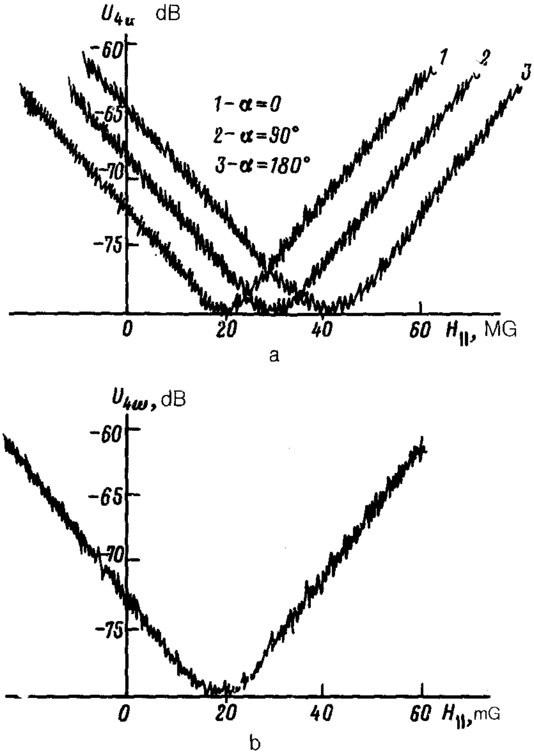

FIG. 2. Amplitude of the fourth harmonic versus the static longitudinal magnetic field $H_{\parallel}$ for a set of single-crystal samples with and without a perpendicular field. a—$H_{\perp} = 0$; b—$H_{\perp} = 2.4$ G. Here α is the angle through which the sample is rotated around the *c* axis from the direction of the magnetic field.

edge effects should have always left the even harmonics with a finite amplitude, i.e., a shift of the "zero" level of the amplitude $U_{l\omega}$. However, we also failed to detect such a level shift.

As Webber *et al.* have pointed out,[5] it might be possible to orient the internal field of the anyons by means of an external magnetic field at temperatures $T_c < T < T_s$. To observe this effect, we carried out the following experiment. An additional magnetic field of 140 Oe was applied in the direction parallel to the axis of the crystals at $T = 150$ K. (According to the data of Ref. 5, we have $T_s - T_c = 40$ K for Bi samples.) The sample was then cooled to 115 K. The external field was then reduced to zero, and the sample was cooled further to $T = 77$ K. At this temperature, we carried out a cycle of measurements, as described above. The results were again negative.

In summary, these experiments allow us to assert that if an internal field of anyons does exist in the high-T_c superconductors, then its magnitude is less than 10^{-3} G.

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