Detection of quantized flux penetration into very small 0-type Josephson links in $Bi_2Sr_2Ca_1Cu_2O_y$ single crystals using ESR spectrometry

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Magnetic-flux trapping in $Bi_2Sr_2CaCu_2O_y$ single crystals is investigated using an electron-spin-resonance spectrometer to study the trapping mode of high-T_c superconductors. Several series of periodic microwave (MW) absorption spectra were observed at external fluxes $\Phi_x=(k\pm 1/2)\Phi_0$, $k=0,1,2,\ldots$. They are superposed on a broad MW spectrum ascribed to the surface impedance. The periodic spectrum is clear evidence for stepwise transitions $(k\rightarrow k+1)$ between energy states of Josephson-junction links by microwave absorption and for stepwise penetration of flux quanta Φ_0. The periodicity with a half quantum $(k\pm 1/2)\Phi_0$ was shown to be due to 0-type Josephson-junction links. It was possible to follow the angular dependence of a few sets of periodic spectra and to determine the sizes and orientations of the intracrystal Josephson links. The link size and the number of quanta penetrating the links are very small, an order of 0.5–4 μm^2 and several to a few tens of quanta, and are about $10-10^2$ times smaller than those in Y-Ba-Cu-O and $Tl_2Ba_2CaCu_2O_8$ crystals. Every link surface is found to be closely located in the *ab* crystal plane. [S0163-1829(97)05521-5]

I. INTRODUCTION

The study of the magnetic-flux-trapping modes in single crystals of ceramic superconductors is important in understanding high-T_c superconductivity and in applications, since flux trapping governs the conductivity in magnetic fields. There may be superconducting loops containing intracrystal Josephson junctions (JJ's) such as the superconductor/(dislocation)/superconductors, which are created by dislocation, in addition to the intracrystal JJ's formed by the intrinsic insulator layers in high-T_c superconductors as well as Abrikosov vortices. These loops play an important role in flux trapping. There are several techniques for studying flux-line lattices (FLL) and the flux quantum Φ_0. Each method has its advantages and limitations. Neutron-scattering probes the bulk features of the FLL.[1] The Bitter decoration method using scanning electron microscopy or atomic-force microscopy is more powerful in studying the details of the structure and the pinning of the FLL.[2,3] However, it probes the FLL mode only at a weak magnetic field of $H_0<500$ Oe and does not provide direct information on Φ_0. The microwave (MW) magnetoabsorption measurement using electron-spin-resonance (ESR) spectrometers directly probes the flux quantum Φ_0 trapped in Josephson junction links,[4–12] as described later. Since JJ links have multienergy (discrete eigen) states, the measurements give rise to an equally spaced line spectrum as the results of transitions between the successive eigenstates by microwave absorption.[4,5,8–12] The periodic magnetoabsorption spectrum is clear evidence for the stepwise penetration of quantized flux into JJ links and provides fundamental flux-trapping information, such as the size and orientation of the Josephson links,[5,9–12] to a high field of the limit of the ESR spectrometers $H_0=5000$–20 000 Oe. On the other hand, the Abrikosov vortices do not give such a periodic spectrum but give a structureless broad spectrum due to microwave surface impedance.[13–17] Here the microwave response of $Bi_2Sr_2CaCu_2O_y$ (Bi2212) single crystals is studied, however, the resolved spectrum due to Φ_0 is not found,[18] in contrast to observations for Y-Ba-Cu-O,[5,7–10] Tl2212,[11] and $ErBa_2Cu_3O_y$ crystals.[12] The Bi2212 superconductor shows a lower magnetoresistivity than the Y-Ba-Cu-O and Tl2212 superconductors at high temperatures.[19–21] To understand the different trapping modes affecting the resistivity we have studied Bi2212 single crystals using a previously improved ESR spectrometer.[20]

II. MICROWAVE RESPONSES FROM JUNCTION LINKS

The MW magnetoabsorption spectrum can be measured using an ESR spectrometer by sweeping the static magnetic field. A few sets of periodic line spectra are observed for Y-Ba-Cu-O and Tl2212 single crystals.[5,7–10] The structure and the angular dependence of the spectra have been reasonably understood as a microwave absorption by JJ links. The spectra reported are composed of periodic lines at a series of the magnetic fluxes $\Phi_x=(k+1/2)\Phi_0$, instead of a periodicity of $\Phi_x=k\Phi_0$, $k=$integer.[5,9–11] The cause of the periodicity including a half quantum $(1/2)\Phi_0$ is interesting in relation to d-wave mechanism of superconductivity.[22–25] Therefore we presently discuss the difference between the responses from 0 and π junctions, which may relate to the periodicity.

The flux states of a Josephson loop or link are discussed theoretically by Silver and Zimmerman,[4] and others.[5–11] The essential ideas are given briefly as follows. The supercurrent I and the Gibbs' free energy E of JJ links are given by

$$I=I_c\sin(\Delta\phi), \tag{1}$$

$$E=LI^2/2-(\Phi_0 I_c/2\pi)\cos(\Delta\phi), \tag{2}$$

where Φ_0, I_c, L, and $\Delta\phi$ are the flux quantum, critical current, inductance of the loop, and the supercurrent phase shift across the Josephson junctions, respectively.[4] For the cases $I_c>0$ and $I_c<0$, the energy of the JJ links has minimum value at the phase shift $\Delta\phi=0$ (exactly, $0+2k\pi$, k indicates integer) and $\Delta\phi=\pi$ (exactly, $\pi+2k\pi$), respectively, as is seen from Eq. (2). The JJ links have multieigenstates E_k with a phase shift by 2π between k and $k+1$ states. The above two junctions, respectively, are named 0 junctions and π junctions.[4,22–25] Whether the real junctions in superconductors belong to the 0 junction or π junction depends on the superconductivity mechanisms concerning s-wave and/or d-wave spin-pairing models and on the relative orientation of the crystallographic axes with respect to the Josephson junction interface.[22–25]

Generally for links containing a number M of junctions, the phase shift is given by Eqs. (3) and (4) from the quantization condition of the phase integral around the link, which leads to the flux quantization inside the links,[5]

$$\Delta\phi=(2\pi/M)(k-\Phi/\Phi_0) \quad \text{for 0 junctions,} \tag{3}$$

$$\Delta\phi=(2\pi/M)(k+M/2-\Phi/\Phi_0) \quad \text{for } \pi \text{ junctions,} \tag{4}$$

where Φ is the flux penetrated across the link surface S. It should be noted again that the JJ links have multieigen states E_k, which have a phase shift by 2π between k and $k+1$ states but have an equivalent energy, as is seen from Eqs. (2)–(4). Namely, the links can accept a number of fluxes by changing the phase shift by 2π, which differs from Abrikosov vortices, as described later.

The shift $\Delta\phi$ for an even number ($M=2m$) of π junctions reduces from Eq. (4) to Eq. (3) by replacing $k\rightarrow k+m$. For an odd number ($2m+1$) of π junctions, it becomes

$$\Delta\phi=(2\pi/M)(k'+1/2-\Phi/\Phi_0) \quad \text{for odd } \pi \text{ junctions.} \tag{5}$$

Hereafter we call the links containing 0 junctions, or an even number of π junctions, 0-type links, and those containing an odd number (including one) of π junctions, π-type links. Using Eqs. (2), (3), and (5), the energy of the links is shown in Figs. 1(a) and 1(c) for 0-type,[4] and π-type links,[22] respectively, as a function of the flux Φ_x due to the external magnetic field $\mathbf{H}_x$,

$$\Phi_x=\Phi_{dc}(t)+\Phi_\mu \sin\omega t, \tag{6}$$

$$[\mathbf{H}_x=\mathbf{H}_{dc}(t)+\mathbf{H}_\mu \sin\omega t],$$

where $\Phi_{dc}(t)$ and $\Phi_\mu \sin\omega t$, respectively, denote the fluxes due to the dc magnetic field $\mathbf{H}_{dc}(t)$ which is swept in ESR measurements and the microwave magnetic field $\mathbf{H}_\mu \sin\omega t$ oscillating at an angular frequency ω. The flux Φ_m due to the modulating magnetic field $\mathbf{H}_m$, which is applied in the ESR measurements, is omitted in Eq. (6), since it acts only as the reference for detecting the MW response in the first derivative form via lock-in amplification if a very small H_m is used: $H_m \ll H_{dc}(t)$.[10,11] The energies have minimum value at $\Phi_x=k\Phi_0$ for 0-type links and $\Phi_x=(k+1/2)\Phi_0$ for π-type links as shown in Figs. 1(a) and 1(c).

In the case of large value for LI_c or M described by the condition $2LI_c+M\Phi_0/2>\Phi_0$, two adjacent energy states $E_k(\Phi)$ and $E_{k+1}(\Phi)$ overlap [Figs. 1(a) and 1(c)].[4,5,22] Then a resonant transition between $E_k(\Phi)$ and $E_{k+1}(\Phi)$ states occurs by application of microwave magnetic field, and as-

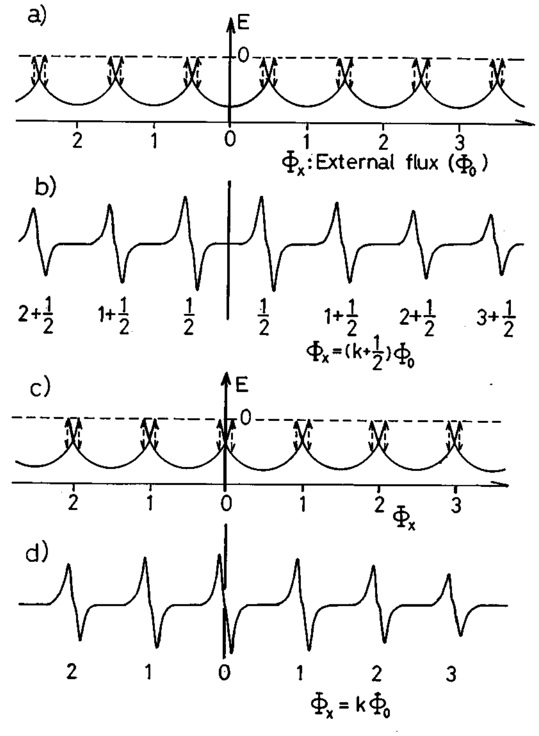

FIG. 1. Energy states of a superconducting link having (a) a 0-type and (c) a π-type Josephson junction. (b) and (d) are microwave spectra due to the transitions (vertical arrows) between states for (b) 0-type and (d) π-type Josephson links. The energies are shown for the condition $\Phi_0/2<M\Phi_0/4+LI_c$ for the number M of the Josephson junctions in the loop (Refs. 4 and 5).

sociating penetration of a flux quantum Φ_0 into the link for each transition, as increasing the external field H_x.[4,5] If we applied a lower MW magnetic field $H_\mu \sin\omega t$ than the dc field H_{dc} (except for near-zero dc field), the MW field acts approximately only as an oscillating field to produce the transition and the external field H_x is approximated to be the dc magnetic field H_{dc} (therefore $\Phi_x=\sim\Phi_{dc}$ for flux). As a result, a periodic MW absorption spectrum with a dc-field separation ΔH, which corresponds to Φ_0, is observed by sweeping the dc magnetic field H_{dc} using an ESR spectrometer, as shown in Fig. 1. When a high microwave power is applied at near zero or small dc field the microwave currents in Josephson junctions also cause fluxon nucleation and annihilation and give rise to a small additional splitting.[5,7,9] Here we do not discuss the additional splitting since the splitting was not clearly observed for the present Bi2212 crystals probably because of the larger dc field than the applied microwave field even for the first line of the periodic spectrum: $H_{dc}/2$(~several to several tens Oe)$\gg H_\mu$(~0.01–0.1 Oe). In the case of the condition $2LI_c+M\Phi_0/2<\Phi_0$, the two adjacent states do not overlap, and result in no resonant transition by microwave field and hence give no MW absorption with a periodicity.[4] Anyhow the observation of a periodic MW response is clear evidence for stepwise penetration of flux quanta into JJ links.

The transitions occur at a series of external fluxes shifted by $\Phi_0/2$ from the energy minimum position as shown in Fig. 1. So the transitions will be observed stepwise at the external fluxes $\Phi_x=(k+1/2)\Phi_0$ for 0-type links and $\Phi_x=k\Phi_0$ for

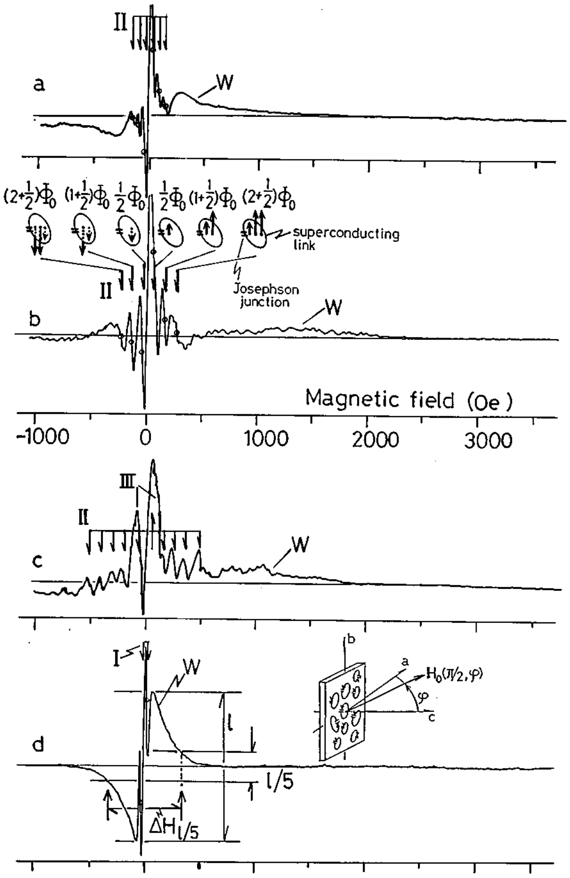

FIG. 2. Microwave spectra observed at 77 K for a thin Bi2212 single crystal (i) of a size $a\times b\times b\times c=1.2\times4.0\times0.008$ mm^3 using an ESR spectrometer by applying the magnetic field parallel to the directions making angle $\Psi=$(a) 70°, (b) 75°, (c) 82.5°, and (d) 0° with the c axis in the ac crystal plane. The modulation frequency is 100 kHz and the modulation width 0.05 Oe. The structures designated I–III originate from flux quanta Φ_0 penetrating into Josephson links. Successive penetration at the external fluxes $\Phi_x/\Phi_0=\pm1/2,1\pm1/2,2\pm1/2\ldots$ indicates that the links are 0 type.

π-type links. Our suggestion is that by this relation we can identify the 0-type and π-type links by detection or by nondetection of the microwave absorption spectrum due to transitions at external fluxes involving the half-quantum $\Phi_0/2$, as shown in Figs. 1(b) and 1(d).

On the contrary, although there exist a large number of Abrikosov vortices, they do not give rise to a periodic MW spectrum. Each vortex accompanies a single flux quantum,[26] namely, the energy of the Abrikosov vortices is single valued. Therefore Abrikosov vortices cause only a structureless magneto-MW response due to microwave surface impedance relating fluxon nucleation,[13–17] and never give rise to a periodic MW absorption spectrum which is characteristic of multivalued energy states. Thus we can selectively extract the information about JJ links by detecting the periodic MW spectrum using ESR spectrometer.

III. EXPERIMENTS

A crystal of $Bi_2Sr_2CaCu_2O_y$ superconductor (size: ~5.3 mm$^2\times20$ mm) was grown by the floating-zone method. Several sample crystals of different thickness were

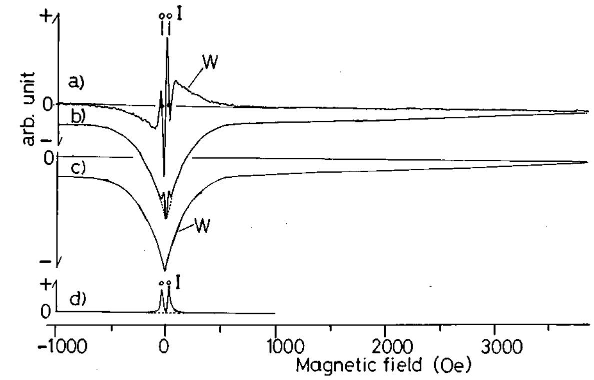

FIG. 3. Microwave spectra in the magnetic-field directions making angle $\Psi=50°$ in the ca plane for a Bi2212 single crystal (i); (a) the first derivative spectrum measured, (b) spectrum in absorption form obtained by integration of (a), and the component spectra of (c) I and (d) W components (see text).

prepared by cutting and cleaving. Every sample was confirmed by x-ray diffraction to be a single crystal. Measurements of the microwave response for those crystals were made at 77 K using a JEOL-FGX3 ESR spectrometer with 100 kHz field modulation. A small modulation field of $H_m\sim0.01-0.1$ Oe is used in the measurements so as to minimize the effect in flux trapping as is discussed in the previous section ($H_m\ll H_0$, except for zero field $H_0=0$). We have improved previously the magnetic power supply of the spectrometer so as to sweep a wide range of the static magnetic field, across zero: $-1000-+9000$ Oe.[20,21] The magnetic-field strength was measured using a gauss meter, Denshijiki Ind. Co., Ltd. GM-003. The microwave power applied is 0.1–1 mW. The MW magnetic field H_μ is crudely estimated to be an order of 10^{-3} Oe, which is much smaller than the dc magnetic field.

IV. RESULTS AND DISCUSSIONS

A. Microwave responses

Figure 2 shows the microwave response spectra in the first derivative form observed at 77 K for a thin sample, denoted (i), having a size $a\times b\times c=1.2\times4.0\times0.008$ mm^3. The spectra (a)–(d) are measured by applying the magnetic field parallel to the directions making angle $\Psi=$(a) 70°, (b) 75°, (c) 82.5°, and (d) 0° with the c axis in the ca crystal plane [Ψ is defined in the inset in Fig. 2(d)]. Well-resolved and equally spaced lines originating from flux quanta were observed for Bi2212 using thin single crystal, denoted by I, II, and III in Fig. 2. They are superposed on a broad spectrum denoted W, which is supposed to be the microwave surface impedance.[13–17] The periodic spectra I–III seem to have opposite polarity to the broad component W as is seen from Figs. 2(a)–2(d). We integrated the spectra to confirm the polarity and to get the real resonant magnetic fields of the lines. The spectra (a) and (b) in Fig. 3, respectively, show the first derivative spectrum observed in the dc magnetic-field direction $\Psi=50°$ in the ca crystal plane and the integrated spectrum (absorption form). Figures 3(c) and 3(d) show the decomposed spectra of Fig. 3(b) and, respectively, present the sharp component II and broad component W. They

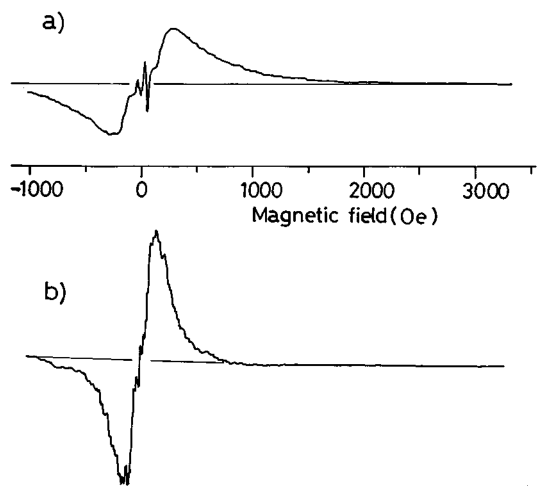

FIG. 4. Microwave spectra observed at 77 K for a Bi2212 single crystal (*ii*) of a size $1.2\times5.7\times0.027$ mm^3 having slightly larger thickness than (*i*). The measurements of the spectra (a) and (b), respectively, are made by applying the dc magnetic field parallel to the same direction as the spectra (b) and (d) in Fig. 2 measured for crystal (*i*). The ESR measurements are made using the same parameters as those for Fig. 2 apart from the amplification gain.

clearly indicate that the two components have opposite polarities, one being 0° phase MW response and the other being 180° (π) phase response to the input phase of MW supplied into the resonant (ESR) cavity. The wind spectrum W in the absorption form has a similar shape to the microwave surface impedance reported,[16,17] confirming that W is due to surface impedance, which is ascribed to the nucleation of Abrikosov fluxons. The cause of the phase difference between the two component responses is not clear at present.

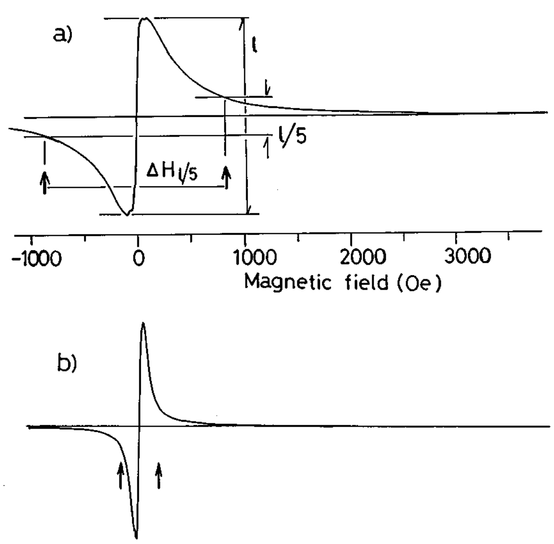

FIG. 5. Microwave spectra observed at 77 K for a thick Bi2212 single crystal (*iii*) having a size $2.6\times3.0\times0.4$ mm^3 for the magnetic field aligned along (a) the a-axis and (b) the c-axis directions. The measurements were made with the same parameters as Fig. 2, apart from the amplification gain. The structures originating from the flux quantum Φ_0 are smeared out by superposition of a number of spectra due to slightly differing Josephson links.

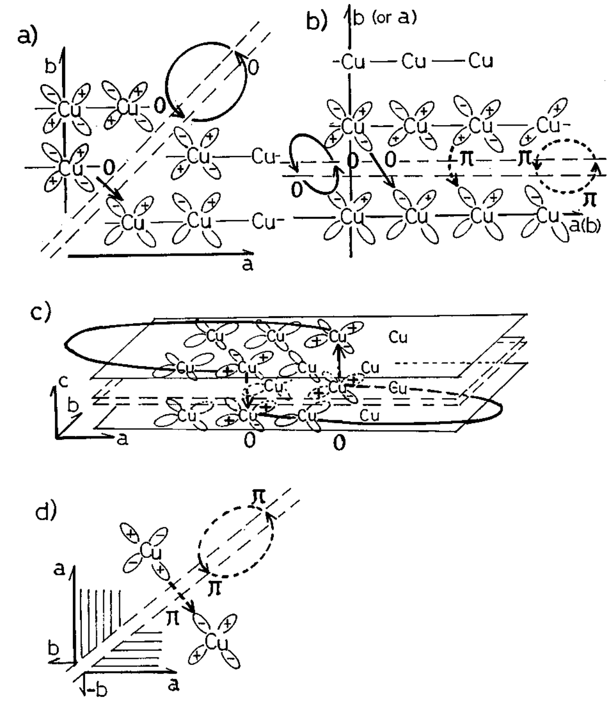

FIG. 6. Illustration of some intracrystal Josephson junction interfaces created by dislocations (broken lines) including (a) the bisector direction of the a and b crystal axes, (b) a- or b-axis direction and (c) c-axis direction, and (d) a twin-type interface. Only d orbitals of Cu^{2+} ions of Bi2212 are shown, since d-wave conductivity mechanism is suggested for cuprate superconductors. The phase change of the supercurrent across the interface is $\Delta\phi=0$ for (a) and (c), π for (d), and 0 or π for (b) depending on the degree of the atom displacement along the dislocation. The resultant phase change along links denoted by thick solid or dotted circles becomes $\Sigma\Delta\phi=0$ or 2π by crossing twice the junction interface.

The structure and the broadness of the spectrum differed from crystal to crystal and depended on the thickness of the crystals. As shown in Figs. 4(a) and 4(b), another crystal having a slightly larger thickness (*ii*; $1.2\times5.7\times0.027$ mm^3) than crystal (*i*) gives broader and more complex spectra in the same magnetic-field orientations as Figs. 2(b) and 2(d), respectively, for the latter crystal. Much thicker crystals such as (*iii*) having a size $2.6\times3.0\times0.4$ and (*iv*) $1.2\times4.0\times2.0$ mm^3 give only structureless broad spectra, as is shown in Fig. 5 for sample (*iii*). These results indicate that there are many sets (groups) of ensembles of JJ links with different sizes and orientations and that the number of the sets increases with the increase of the thickness of the crystal (crystal size). As a result the structures are diminished by superposition of a number of different periodic spectra and disappear into broad component of surface impedance. This might be the cause of the failure in detecting periodic spectra for Bi oxide superconductors in the past ESR study probably using thick crystals.[18]

B. 0-type Josephson links

The successive lines of II are observed at field increments $\pm1/2, \pm(1+1/2), \pm(2+1/2), \ldots$ from the zero field $H_0=0$ as shown in Figs. 2(a)–2(c). This indicates that the transition between states occurs at a stepwise progression of the exter-

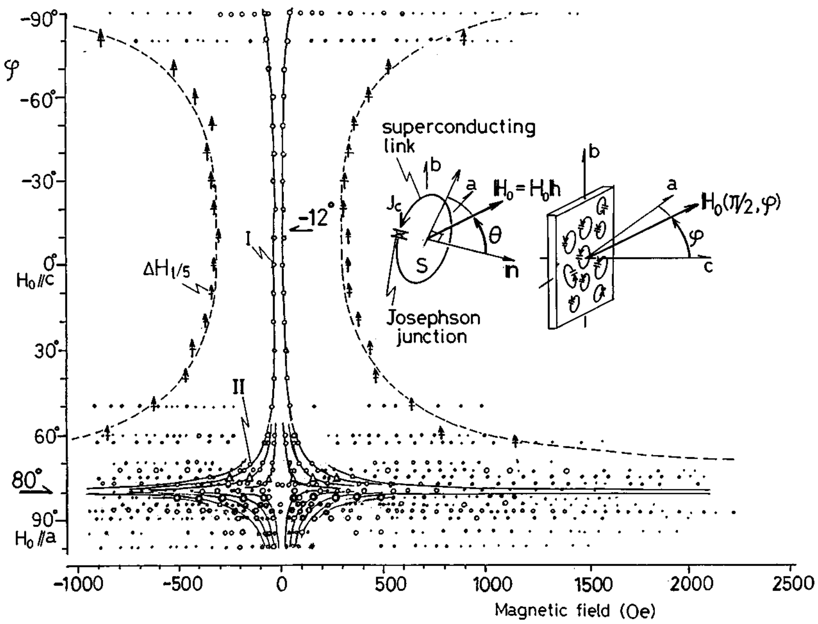

FIG. 7. Angular dependence of the microwave spectra in the *ca* crystal plane for the thin Bi2212 single crystal (*i*). The data points denoted by larger size of circles ○ represent the stronger spectrum line. The triangle △ at an experimental angle $\Psi=75°$ denote the line positions of the periodic spectrum II in Fig. 2(b). The solid curves for spectra I and II show the calculated $1/\cos\theta$ dependence. The spectrum I shows the minimum line-separation at $\Psi=-12°$, which corresponds to the direction of the normal axis **n** of the Josephson link surface projected to the *ca* plane. The spectrum II (and the line separations) diverges at $\Psi=80°$, and indicates that the normal axis makes angle $90°-80°=10°$ with the *c* axis. The inset shows the superconducting links having a Josephson junction and surface area *S* (see text).

nal magnetic flux $\Phi_x=(k\pm1/2)\Phi_0$ [characteristic of 0-type links in Figs. 1(a) and 1(b)] and not the progression $\Phi_x=k\Phi_0$ [characteristic of π-type links in Figs. 1(c) and 1(d)]. The periodicity of $(k\pm1/2)\Phi_0$ is also reported for Y-Ba-Cu-O and Tl2212 single crystals.[5,11] Thus it may be concluded that the intracrystal links in the single crystals of these ceramics superconductors contain 0 junctions or an even number of π junctions, and do not contain a single (or an odd number of) π junction.

This might be understood by intracrystal Josephson junctions and JJ links illustrated in Figs. 6(a)–6(d), where the links are located in or near the *ab* plane since the location was experimentally suggested as described later. The cases (a)–(d) present some possible interfaces created by dislocations involving (a) the bisector direction of the *a* and *b* crystal axes, (b) the *a*- or *b*-axis direction and (c) the *c*-axis direction, and (d) a twin-type interface. The phase change of the supercurrent across the interface is $\Delta\phi=0$ for cases (a) and (c), π for (d), and 0 or π for (b) depending on the degree of the displacement of atoms along the dislocation. However, the current twice crosses the junction interface along the link circuit in every case, as indicated by thick solid or dotted loop. Even in a case such as (b), the penetration of supercurrent to the junctions will occur along the loop with the phase changes 0-0 or π-π rather than a loop with the changes 0-π or π-0, since the 0 or π junction has a lower tunnel barrier than the other. As a result, the resultant phase change becomes $\Sigma\Delta\phi=0$ or 2π, and interpreting the above experimental evidence of the periodicity $(k\pm1/2)\Phi_0$.

C. Size and orientation of JJ links

We measured a series of MW-response spectra such as Fig. 2 for the sample (*i*) by rotating the crystal in an interval of the rotation angle $\Delta\Psi=5°\sim10°$ with respect to the external magnetic field ($\mathbf{H}_0$) direction in the ESR cavity. The spectral line positions are plotted as a function of the experimental rotation angle Ψ. Figure 7 shows the angular dependence of the spectra observed in the *ca* crystal plane. Figure 8 is essentially the same angular dependence as Fig. 7, but is an expanded (precise) plot in a range of angle $\Psi=62.5$–$95°$ for the spectra, which are measured in a smaller angle interval $\Delta\Psi=2.5°$. Even for the thinnest crystal (*i*) of the present samples, the MW response spectrum was found to be composed of many sets of periodic lines with different spacings and different angular dependences. The spacing ΔH between successive equally spaced lines varied like a function of $1/\cos\theta$ as well as the magnetic-field position of each line, where θ is a direction angle described later. We were able to follow the angular dependence only for three periodic spectra which are denoted by I, II, and III in Figs. 7 and 8, where the solid curves denote the $1/\cos\theta$ dependence. It was impossible to follow other weak lines because there were so many of them. The present observation of periodic spectra along with the angular dependence in the $1/\cos\theta$ form is clear evidence for the stepwise penetration of quantized fluxes into JJ links, as discussed in Sec. II.

The line separation is given by $\Delta B=\Phi_0/S_J$, since one flux quantum Φ_0 penetrates into the link surface S_J in each increment of the flux density ΔB which is due to the increment of the magnetic field ΔH.[5,10,11] The separation shows an angular dependence of $1/\cos\theta$. This is because the effective surface $S_{\rm eff}$, into which the fluxes penetrate, is changed by the external field direction ($\mathbf{h}=\mathbf{H}_0/H_0$). $S_{\rm eff}$ is given by the cross section of the surface S_J (having normal **n**) projected onto the plane normal to the field direction **h**: $S_{\rm eff}$

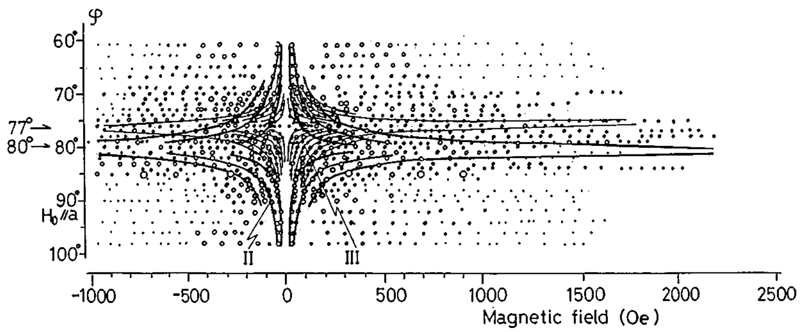

FIG. 8. Expanded plot of the angular dependence of the microwave response spectra in Fig. 7 around the a axis in the ac crystal plane. The spectra II and III (and the line separations), respectively, diverge at $\Psi=80°$ and 77°, and indicate that the normal axis of the Josephson links make angles 10° and 13° with the c axis in the ca plane (see text).

$=\mathbf{h}\cdot\mathbf{n}S_j=S_j\cos\theta$, where θ is the angle between **h** and **n** as shown in the inset in Fig. 7.[5] ΔB is given by

$$\Delta B=\Phi_0/S_j\cos\theta. \tag{7}$$

The spacing ΔH and the response spectrum diverge at $\theta=90°$ and have minimum values at $\theta=0°$, which corresponds to the normal axis direction of the JJ link surface. One needs the angular dependence of the spacing in the three crystal planes (ca, ab, and bc) to determine the direction cosines ($\sin\zeta\cos\xi$, $\sin\zeta\sin\xi$, $\cos\zeta$) of the normal axis **n** in the polar coordinates (ζ, ξ) of the a, b, and c axes. Figures 7 and 8 show that the normal axes **n** for I, II, and III made angles (−)12°, 10°, and 13° with the c axis in the ca plane. The results indicate that **n** are closely located to the c-axis direction, i.e., the surface plane of the links being in the ab plane. As shown in Fig. 7, other weak lines are observed around the a-axis direction and not observed around the c direction, although they were difficult to follow the angular dependence. The results suggest that the link surface planes for these weak lines are also closely located in the ab plane.

It was unfortunately also difficult to follow the angular dependence of the spectra including I, II, and III in the ab and bc planes since poorly resolved spectra were observed. Especially, as shown in Figs. 9(a)–9(d), the MW responses in any direction in the ab plane showed broad and widely spread spectra superposing many weak lines on the broad component. These results, however, lend support to the claim that all the surface planes of links (probably including I–III) are closely in the ab plane since the spectra are spread when the magnetic field is applied on the link surface planes.

Similar periodic spectra have been reported for Y-Ba-Cu-O,[5,7–10] Tl2212,[11] and $ErBa_2Cu_3O_y$ single crystals.[12] However, the spacing observed for Bi2212 is much larger, by about 10^2 times, than those reported for the latter compounds. From the angular dependence in the ca plane (Figs. 7 and 8), the link surfaces projected on the ab plane (along the c axis) are estimated to be $S_J=0.5\pm0.1$, 3 ± 1 and 4.4 ± 0.3 μm^2 for I, II, and III, respectively. The real surfaces will not differ much from these values since their surface planes were suggested to be closely in the ab plane. The link sizes are extremely small compared to the sizes $S_J=160$ and 804 μm^2 reported for Y-Ba-Cu-O,[5] and $S_J=211$ μm^2 for Tl2212 single crystals.[11] For spectrum I this might be due to Abrikosov vortices rather than JJ links, since it does not give a periodic spectrum but rather only one line for ±magnetic-field sides. If it were the case, the effective loop size for I should be multiplied by factor 2 ($0.5\times2=1$ μm^2) since the fluxon nucleation and annihilation occur at a field corresponding to $\pm1\Phi_0$ rather than $\pm(1/2+k)\Phi_0$.

The number of the periodic lines ranges from several to a few tens as is seen for II, III, and other weak lines in Figs. 2, 7, and 8. This differs from the several tens and hundreds reported for Y-Ba-Cu-O and Tl2212 compounds.[5,7–11] The present smaller values may be ascribed to a limit of the number of quanta acceptable to the Josephson links in Bi2212 because of the small link size.

D. Angular dependence of surface impedance

The angular dependence of the overall width of the microwave response spectrum, which is related to the width of the surface impedance spectrum, is also plotted in Fig. 7 with arrows. We defined the width as the separation $\Delta H_{1/5}$ between the field positions giving one fifth of the maximum

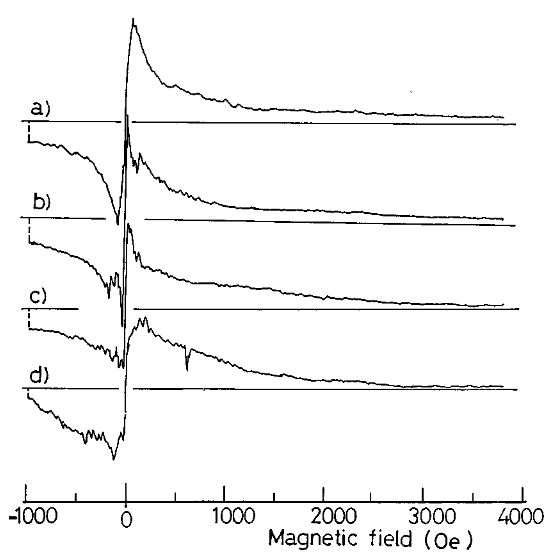

FIG. 9. Microwave spectra observed for a single crystal (i) by applying the magnetic field parallel to the directions making angle Ψ=(a) 0° ($H_0\|a$ axis), (b) 30°, (c) 60°, and (d) 90° ($H_0\|b$ axis) with the a axis in the ab crystal plane.

height of the broad component as shown in Fig. 2(d). It varies like a function of $1/\cos\theta$ as shown with dotted curves in Fig. 7, and as do the periodic lines. The spectrum width of the thick crystals (ii)–(iv) also showed a similar angular dependence to that of crystal (i), and had minimum value in a direction close to the c axis as well as the Josephson links. These results indicate that the nucleation and annihilation of Abrikosov fluxons, which is the origin of the MW surface impedance,[14–16] occur in similar way (with a similar angular dependence) of the quantized flux penetration and annihilation into Josephson junctions for increasing and decreasing the dc field.

The present work indicates that the surface planes of the JJ links in Bi2212 crystals are closely located in the ab crystal plane (the normal axis **n** of the surface being near the c axis). The close location in the ab plane may originate from the higher conductivity in the ab plane than other directions. However, the link surfaces are slightly tilted from the ab plane ($\sim 10°$–$20°$). Therefore the most plausible JJ links might be a loop shown in Fig. 6(c), where the supercurrent flows in two ab planes, which are separated by a JJ boundary created by dislocation parallel to the ab plane, by tunnel crossing the boundary. Then the slight tilt of the link surfaces from the ab plane can be understood. The cause of the small link sizes observed for Bi2212 is not clear at present. One possible speculation might be its lower conductivity than other superconductors, since the lower conductivity might show a tendency to make a smaller superconducting loop. To get further information about the JJ links in high-T_c superconductors it is necessary to prepare artificial superconductive multilayers, artificial Josephson-junction layers, and layers involving insulator particles and to study the microwave response from them. The study in this direction is proceeding.

V. CONCLUSION

The present work has detected the quantized flux penetration into intracrystal Josephson junctions of Bi2212 single crystals. It elucidates a large difference of the flux-trapping mode in Bi2212 from other ceramic high-T_c superconductors. The size and the number of quanta acceptable to the junction links are about $10-10^2$ times smaller than Y-Ba-Cu-O and Tl2212 single crystals.

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