Josephson plasma resonance in the mixed state of the organic superconductor κ-(BEDT-TTF)$_2$Cu(NCS)$_2$

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Magnetic-field dependence of microwave surface resistance measured using a cavity perturbation technique shows a peak structure below T_c in the organic layered superconductor κ-(BEDT-TTF)$_2$Cu(NCS)$_2$. It is observed only when E_ω is perpendicular to the superconducting layer and the peak field shows an anticyclotronic frequency dependence. All these observations suggest the excitation of Josephson plasma across the superconducting layers as the origin of the resonance. The interlayer phase correlation increases as the temperature is lowered even below the irreversibility line in contrast to the case of the $Bi_2Sr_2CaCu_2O_{8+y}$ high-T_c superconductor. [S0163-1829(97)50122-6]

The layered structure of highly anisotropic superconductors leads to some exotic phenomena concerning the interlayer intrinsic Josephson coupling. It is well established that the CuO_2 superconducting layers in high-T_c cuprate superconductors are coupled intrinsically by the Josephson interaction.[1–3] Such an intrinsic Josephson effect has been also reported[4,5] in the layered organic superconducting salt κ-(BEDT-TTF)$_2$Cu(NCS)$_2$, where BEDT-TTF is bisethylenedithio-tetrathiafulvalene.

Recently, the collective Josephson plasma modes excited by electromagnetic waves have attracted much attention in high-T_c superconductors. Because of a strong anisotropy, the plasma frequency ω_p for the polarization perpendicular to the superconducting layers is pushed down to the far-infrared region, in some cases down to the microwave region. This realizes an unusual situation that $\omega_p<2\Delta$, which enables us to observe optically a Josephson plasma mode as an extremely sharp reflectivity edge below T_c in some high-T_c superconductors.[6] Moreover, it is predicted that the Josephson plasma frequency decreases further under a magnetic field.[7,8] In $Bi_2Sr_2CaCu_2O_{8+y}$ (BSCCO) high-T_c superconductors, Tsui *et al.*[9] and Matsuda *et al.*[10] have found the magnetoabsorption resonance in the frequency range of 30–60 GHz. Similar resonance has been observed also in single-layer $Bi_2(Sr,La)_2CuO_y$.[11] These resonances show anticyclotronic behavior and are called ''Josephson plasma resonances.''

In such a Josephson plasma resonance (JPR), the temperature and frequency dependence of the resonance peak can yield information on the interlayer phase coherence under a magnetic field.[8,10] Thus, this kind of measurement is a powerful tool for the microscopic information on the mixed state of layered superconductors. One of the unresolved questions of JPR is the temperature dependence of interlayer phase coherence $\langle\cos\phi_{n,n+1}\rangle$, where $\phi_{n,n+1}$ is the gauge-invariant phase difference between layers n and $n+1$, and $\langle\cdots\rangle$ denotes thermal and disorder averaging. In BSCCO it shows a sharp cusp at the irreversibility line, below which $\langle\cos\phi_{n,n+1}\rangle$ decreases as the temperature is lowered. This means that the phase becomes more incoherent at low temperatures. There is a theoretical consideration[12] which explains this nontrivial result, but this feature seems to be inconsistent with a recent result of the time-dependent Ginzburg-Landau (TDGL) simulation.[13]

Another question is whether the JPR is a unique characteristic for CuO_2 planes or not. It is of great importance to compare it with the superconducting state of other layered superconductors such as the organic BEDT-TTF salts and the artificial multilayer systems.

In this paper, we find that the organic superconductor κ-(BEDT-TTF)$_2$Cu(NCS)$_2$ is another Josephson coupled layer superconductor which shows JPR in the mixed state. Our results clearly demonstrate that JPR is not a peculiar feature of high-T_c superconductors but a common phenomenon for layered superconductors with Josephson interlayer coupling. We also find that the temperature dependence of interlayer phase coherence below the irreversibility line is completely different from that in BSCCO; it increases as the temperature is lowered even below the irreversibility line down to 0.5 K.

Single crystals of κ-(BEDT-TTF)$_2$Cu(NCS)$_2$ were grown by an electrochemical oxidation method.[14] Typical dimensions of the crystal are $1\times0.5\times0.04$ mm^3; the shortest is the direction perpendicular to the conducting layer. We have measured 3 crystals with T_c of 9.0 K and we have obtained similar resonances for all the crystals. The sample was put on the sapphire holder in microwave cavities. We used two TE_{011}-mode cylindrical Cu cavities ($\omega/2\pi=$ 24, and 41 GHz, $Q\sim10^4$) coupled by input and output waveguides. In this study, dc magnetic field up to 90 kOe is applied perpendicular to the conducting layers. The surface resistance R_s was determined by changes in Q value measured using a scalar network analyzer. In contrast to the bolometric technique which has been used for the observation of JPR in high-T_c superconductors,[9–11] the Q measurement technique requires lower incident microwave power. This difference

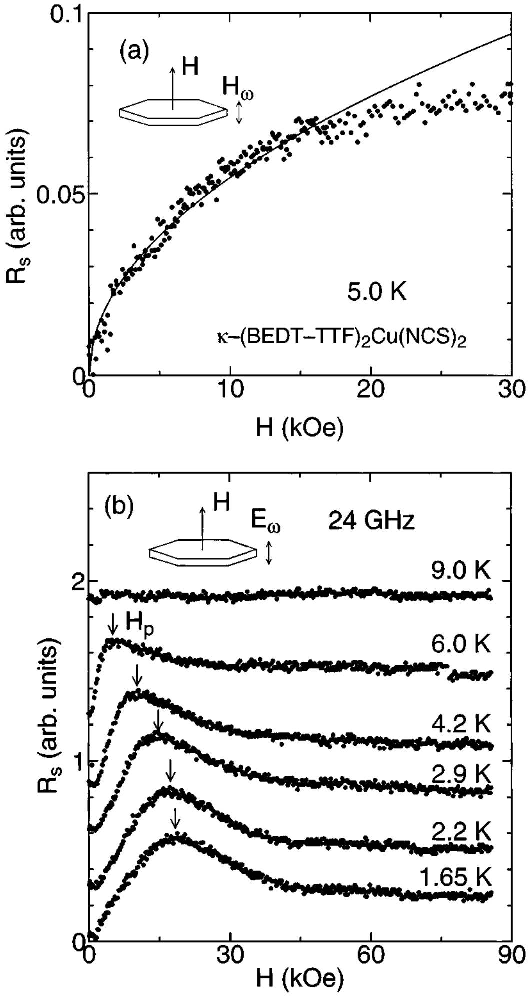

FIG. 1. Magnetic-field dependence of the surface resistance in two microwave field configurations. (a) H_ω is perpendicular to the layer. The solid curve is a $H^{1/2}$ dependence expected in the flux-flow regime. (b) E_ω is perpendicular to the layer. The arrows denote the peak field H_p. Only the increasing field data are shown because no hysteresis was observed.

can be important for materials with low critical current density.[15] In our measurement conditions, the results do not depend on input microwave power (0.1–10 mW) down to ^{3}He temperatures, which ensures that the temperature increase of the sample by microwave absorption is negligible. We also determined the irreversibility line by magnetization hysteresis measurements using a commercial superconducting quantum interference device magnetometer.

Figure 1 shows the magnetic-field dependence of the surface resistance $R_s(H)$ in two microwave field configurations. In Fig. 1(a), the microwave magnetic field H_ω is perpendicular to the conducting layer (b-c plane) and the electric field E_ω is zero. The eddy currents flow in the plane. In this case, vortices are driven by in-plane microwave currents and flux-flow-type dissipation is expected. In fact, R_s is almost proportional to $H^{1/2}$ except for high fields, which is consistent with the flux-flow regime.[16] On the other hand, when the microwave electric field E_ω is perpendicular to the layer and H_ω is zero [Fig. 1(b)], $R_s(H)$ shows a peak at a characteristic field H_p, which is completely different from $H^{1/2}$ dependence. Within our measurement resolution, $R_s(H)$ shows no hysteresis in the increasing and decreasing field branches, which is in contrast to the case in $Bi_2Sr_2CaCu_2O_{8+y}$. This is because the trapped self-field of our crystal is much smaller than H_p and the pinning force or the in-plane critical current density in this compound is much smaller than those in high-T_c superconductors.[17,18] As the temperature is increased, H_p decreases and the peak disappears at $T = T_c$. This result indicates that the peak structure is the response of the superconducting carriers.

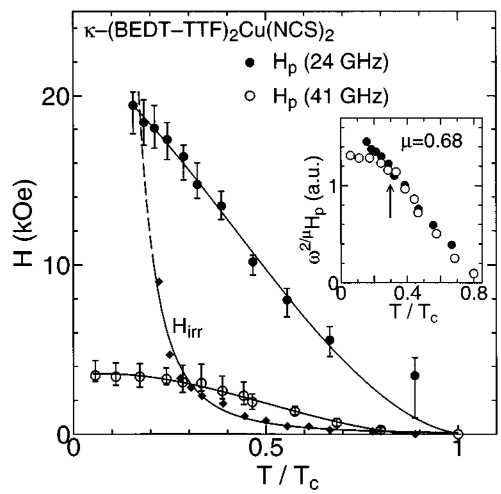

FIG. 2. Temperature dependence of H_p at 24 (closed circles) and 41 GHz (open circles). The irreversibility line (closed diamonds) is also shown. The solid and dashed curves are the guides for the eye. The inset shows $\omega^{2/\mu}H_p$ vs temperature with $\mu = 0.68$. The arrow indicates the crossing point of the irreversibility line and 41 GHz data.

In Fig. 2, we plot H_p at two frequencies (24 and 41 GHz) as a function of temperature. Also shown in Fig. 2 is the temperature dependence of the irreversibility field H_{irr} for another crystal from the same batch, which is consistent with the literature data.[17] It is found that the higher frequency data show smaller values of H_p. This anticyclotronic behavior cannot be explained by the usual cyclotron resonance ($\omega = eH/m^*c$) but is similar to the resonance in BSCCO.[9,10] From the above results together with the polarization dependence in Fig. 1, we assign this peak structure to the Josephson plasma resonance in a magnetic field. The Josephson plasma frequency ω_p is determined by the maximum interlayer Josephson current $J_m(H,T)$:

$$\omega_p^2(H,T) = \frac{8\pi^2 cs}{\epsilon_0 \Phi_0} J_m(H,T), \tag{1}$$

where s is the interlayer spacing, ϵ_0 the high-frequency dielectric constant, Φ_0 the flux quantum. When the magnetic field is applied perpendicular to the conducting layer, J_m and hence ω_p decrease with increasing field. Thus anticyclotronic resonance occurs when the Josephson plasma frequency coincides with the measurement frequencies.

There are, however, considerable differences from the case of BSCCO. In BSCCO, H_p increases as T is lowered above the irreversibility line, and decreases monotonically [$\propto T$ (Ref. 10) or $\propto \exp(T/T_0)$ with $T_0 \sim 12$ K (Ref. 19)] be-

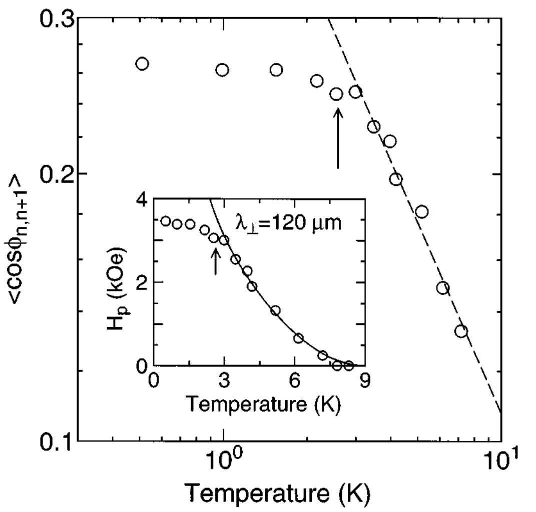

FIG. 3. Temperature dependence of the interlayer correlation $\langle\cos\phi_{n,n+1}\rangle$ at $H=3.46$ kOe extracted from 41 GHz data. The dotted line is the power-law temperature dependence $T^{-0.72}$. The inset is the temperature dependence of H_p at 41 GHz with a theoretical fit of Eq. (3) (solid curve). The arrows indicate the crossing point of the irreversibility line and H_p at 41 GHz.

low that; $H_p(T)$ has a definite cusp at the irreversibility line. In our case of κ-(BEDT-TTF$)_2$Cu(NCS$)_2$, however, there are no sharp cusps in $H_p(T)$ and only the slope dH_p/dT changes at the irreversibility line. To see this more clearly, we plot in the inset H_p data at two frequencies scaled to $\omega^{2/\mu}H_p$, where μ is a scaling parameter. The arrow indicates the temperature where the 41 GHz data cross the irreversibility line. It is evident that the scaled $\omega^{2/\mu}H_p$ below the irreversibility field $H_{irr}(T)$ is suppressed compared with the same temperature data above H_{irr}. This means that the interlayer correlation of vortices definitely changes at the irreversibility line.

According to the recent calculation by Koshelev,[20] the maximum Josephson current in the high-frequency regime[21] can be given by

$$J_m(H,T)=J_0(T)\langle\cos\phi_{n,n+1}\rangle(H,T). \qquad (2)$$

Here the parameter $J_0(T)=c\Phi_0/8\pi^2 s\lambda_\perp^2(T)$ characterizes the Josephson interlayer coupling at zero field, and $\lambda_\perp$ is the out-of-plane penetration depth. In his calculation, the Josephson plasma frequency in the decoupled liquid phase can be written as

$$\omega_p^2=\frac{2\pi f s J_0^2(T)\Phi_0}{\epsilon_0 k_B T H}, \qquad (3)$$

where f is the dimensionless function of order unity with weak temperature dependence. The inset of Fig. 3 shows the theoretical fit of Eq. (3) to the measured $H_p(T)$ at 41 GHz.[22] We have assumed $f=1,\epsilon_0=25$, and Ambegaokar-Baratoff-type temperature dependence of J_0,[23]

$$J_0(T)=J_0(0)\frac{\Delta(T)}{\Delta(0)}\tanh\left[\frac{\Delta(T)}{2k_BT}\right], \qquad (4)$$

where Δ is the superconducting gap. The experimental data above the irreversiblity line are well fitted with the single fitting parameter $\lambda_\perp=120$ μm. It should be noted that this value is close to that obtained in other measurements of the penetration depth.[4] From this value we can estimate the anisotropy parameter $\gamma=\lambda_\perp/\lambda_\parallel$ to be 100–180 taking $\lambda_\parallel=0.65$ (Ref. 24)–1.2 μm.[25] This obtained value is consistent with the published data of $\gamma=120$ (Ref. 26) and 160–350.[4] It demonstrates that this organic superconductor is highly anisotropic and the anisotropy is comparable to that of BSCCO rather than $YBa_2Cu_3O_7$.

By using Eqs. (1) and (2) the interlayer phase coherence can be directly extracted from the temperature and frequency dependence of this resonance peak. The main panel of Fig. 3 shows the extracted temperature dependence of $\langle\cos\phi_{n,n+1}\rangle$ at $H=3.46$ kOe using $\mu=0.68$,[27] which is obtained from the frequency dependence (inset of Fig. 2). We have chosen the field $H=3.46$ kOe so that the result of $\langle\cos\phi_{n,n+1}\rangle(T)$ below the irreversibility line is not affected by the precise value of μ, which is not estimated in the vortex solid state. If we fit the data above the irreversibility line by a power-law temperature dependence $T^{-\beta}$, we get $\beta\simeq0.72$, which is somewhat smaller than that of BSCCO.[10] A more striking difference from the case of BSCCO is that $\langle\cos\phi_{n,n+1}\rangle$ of κ-(BEDT-TTF$)_2$Cu(NCS$)_2$ increases with decreasing temperature even below the irreversibility line. A possible explanation of this temperature dependence is the difference of the pinning strength between BSCCO and κ-(BEDT-TTF$)_2$Cu(NCS$)_2$. The in-plane critical current density of our sample determined by the magnetization measurements is of the order of 10^4 A/cm^2 at low temperatures, which is about 100 times smaller than that of BSCCO. The difference of the pinning force is also visible in $R_s(H)$. In our sample we observed no hysteresis in the increasing and decreasing field branches, while in BSCCO a large hysteresis has been observed particularly at low temperatures.[10] The trapped field is less than the field resolution of typically 100 G, from which we obtain the upper limit of $j_c\sim2\times10^4$ A/cm^2 consistent with the magnetic measurements. When the system has a strong pinning as in the case of BSCCO, vortex displacement from an ideal position becomes large in the case of field sweep measurement, which decreases the interlayer phase coherence. The observed temperature dependence of $\langle\cos\phi_{n,n+1}\rangle$ in κ-(BEDT-TTF$)_2$Cu(NCS$)_2$, however, is consistent with a recent result of the TDGL simulation by Machida *et al.*[13] They obtained the increasing power-law dependence of $\langle\cos\phi_{n,n+1}\rangle$ as T is lowered with different powers for below and above H_{irr} for a weak pinning system, which is in agreement with our experimental result. Therefore we conclude that the pinning strength affects $\langle\cos\phi_{n,n+1}\rangle(T)$ in the vortex solid state. Quite recently, Matsuda *et al.*[28] have shown that $\langle\cos\phi_{n,n+1}\rangle$ does not decrease below the irreversibility line even in BSCCO, when they measure JPR in the temperature sweep condition where the field distribution is more homogeneous.

Turning to the frequency dependence of H_p, we obtain from the inset of Fig. 2 $\omega^2\propto H_p^{-\mu}$ with $\mu=0.68\pm0.1$ above the irreversibility line. The obtained value of μ is smaller than that of BSCCO (0.9−1.1) in the vortex liquid state.[10] According to a recent theory by Bulaevskii, Pokrovsky, and Maley,[12] the exponent μ characterizes ordering of pancake vortices along the direction perpendicular to the superconducting layers and μ increases with disorder. Based upon their theory, our result of smaller μ suggests that this organic superconductor in the vortex liquid phase has weaker disorder in pancake arrangements along the direction perpendicular to the layers than that of BSCCO. The out-of-plane configuration of pancake vortices can be affected by the thermal fluctuations and the pinning strength in each superconducting layer. Thus, the reason for the above difference in μ can be understood considering the differences of the temperature scale and the pinning force discussed in the previous paragraph.

In conclusion, we have observed the peak structure in the magnetic-field dependence of R_s in κ-(BEDT-TTF)$_2$Cu(NCS)$_2$ single crystals. The peak is observed below T_c when E_ω is perpendicular to the layer, and is absent when H_ω is perpendicular to the layer. The frequency dependence of the peak field is expressed as $\omega^2 \propto H^{-\mu}$ with $\mu = 0.68 \pm 0.1$ above the irreversibility line. All these results are consistent with the excitation of the Josephson plasma mode, which is characteristic of the layered superconductors with Josephson coupling. We estimated the out-of-plane penetration depth $\lambda_\perp = 120$ μm yielding the anisotropy parameter $\gamma = 100-180$, which is comparable to the value of BSCCO. The temperature dependence of the peak field below the irreversibility line is completely different from that of BSCCO, implying that the strength of the pinning force strongly affects the interlayer phase coherence in the vortex solid state.

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