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Observation of Al_2O_3:Cr^{3+} magnetic resonance via solitons in long Josephson junctions

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We have detected the zero-field electron-spin resonance signal of Al_2O_3 with ~1000 ppm Cr^{3+} using an edge Josephson junction. The sample comprises the substrate upon which the junction was fabricated and the coupling to the electron-spin resonance is via the solitons which exist in these "long" devices.

Some time ago Barnes[1] suggested that the ac Josephson effect might be used as a method of performing *in situ* electron-spin resonance (ESR). Later Pellisson, Delescelfs, and Barnes[2] demonstrated that suitable superconductor-normal metal-superconductor (SNS) junctions could be fabricated in which the "sample" comprised the N layer in a new type of point-contact junction. Based upon this work, Bures and co-workers[3] were the first to perform successful ESR experiments on Au:Gd^{3+} and Au:Er^{167}. Recently Goldman, Kuper, and Valls[4] and Barnes and Mehran[5] have shown how the same technique might be extended to make q- and ω-dependent determinations of the full dynamical susceptibility $\chi(\mathbf{q},\omega)$. While it has been argued[5] that this technique, like many Josephson-based techniques, might be limited in sensitivity by only intrinsic limitations, it is far from clear that it represents a realistic alternative technique for performing ESR.

In this Rapid Communication we show that it is possible to easily detect the ESR signal of ~1000 ppm of Cr^{3+} in corundum using a conventional edge junction fabricated *on* the sample. Also new is the fact that the ESR is detected via the coupling of the solitons to the magnetic system. This demonstration is important because it shows for the first time that this technique can be applied to materials without the need to incorporate the sample into the junction itself and that the coupling to solitons is possible.

The Cr^{3+} ion in corundum has a narrow *zero-field* ESR transition[6] at 11.447 GHz which via the Josephson relation 2 eV $=h\nu$ corresponds to a voltage of 23.667 μV. It is this same transition which is used in a ruby maser.

We have used a "long" Josephson junction. When the length L exceeds the Josephson penetration length $\lambda_J \equiv (\hbar/2ed\mu_0 J_c)^{1/2}$, where J_c is the critical current density and d is the magnetic thickness of the insulating (I) layer, the basic excitations of a simple superconductor-insulator-superconductor junction change from being the Fiske (or Eck) modes considered in the earlier theories to solitons.

The basic excitation consists of a single soliton, or vortex, which propagates along the junction with a velocity u determined by the average voltage $\bar{V}$ via $\bar{V}=\Phi_0 u/L$, where $\Phi_0 \equiv h/2e$ is the flux quantum. In zero external magnetic field the soliton is reflected at the end of the junction as an antisoliton, i.e., a flux vortex with the magnetic field in the opposite sense. A second reflection recovers the original vortex. Thus, the period $T=2L/u$ and

$$h\nu = h/T = hu/2L = \Phi_0 eu/L = e\bar{V} ,$$

i.e., the relation between the frequency of the magnetic field and the dc voltage is $h\nu = e\bar{V}$, which differs from the usual Josephson relation by a factor of exactly 2. The largest voltage associated with this basic excitation is obtained by observing that the soliton cannot travel faster than the speed of light *in the junction*, $\bar{c}=(2a/d)^{1/2}c$, where $2a$ is the thickness of the I layer and the magnetic thickness $d=2a+\lambda_L+\lambda_L'$ where λ_L and λ_L' are the London penetration depths of the two S layers. The corresponding voltage is $V_{n=1}=\Phi_0\bar{c}/L$. In addition to the branch associated with this simplest excitation are branches which asymptotically reach $V_n=n\Phi_0\bar{c}/L$ and are associated with n solitons propagating along the junction. These voltages V_n correspond to the position of the *even* order Fiske modes. A typical set of such branches is shown in Fig. 2(b).

The junctions were fabricated using a process developed for Nb:Pb alloys based on seven-level integrated circuit photolithographic methods.[7] The junction illustrated in Fig. 1 has the length L determined by the thickness of the lower superconducting film and a width W. The I layer makes an angle of roughly 45° to the substrate which means that the flux which passes through the junction also passes through the substrate, i.e., with such a system there is essentially no difference between having the ESR ions situated in the substrate or within the magnetic thickness of the junction. We will analyze the system as if the latter were the case.

The resonance of Cr^{3+} has been detected in a some-

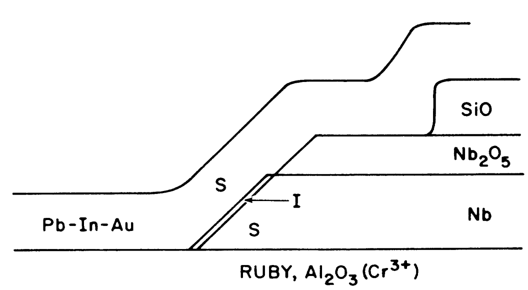

FIG. 1. The edge-junction geometry.

what different fashion in two different junctions. The junctions are (current) fed from a voltage source via a 1-kΩ resistor. A small field ~1 G is applied in the plane of the substrate using a pair of coils.

In one junction (see Table I for parameters) the signal at ~24 μV appears as a vertical step in a soliton branch; this is shown in Fig. 2(a). The feature is very similar to the microwave induced steps. For another junction, fabricated on the same sample, a quite separate branch associated with the resonance appeared *but only in finite, small, magnetic fields* ($H \sim 0.1-1$ G) and now at 47 μV, i.e., twice the voltage predicted by the Josephson relation. The relevant branch is labeled *A-B* in Fig. 2(b). In both cases, the position and height of the steps on the current axis *but not* the position on the voltage axis was sensitively dependent upon the field. In addition to these expected steps, the former junction exhibited a doubling of some of the soliton branches which we also associate with the magnetic substrate [Fig. 2(c)]. One branch is simply a replica of the other displaced along the voltage axis by 47 μV.

These observations can be explained by adding the magnetic coupling to the theory of resistively damped solitons. In the absence of either resistive or magnetic damping it is easy to combine Maxwell and Josephson equations to obtain the sine-Gordon equation,[8]

$$\frac{\partial^2\phi}{\partial x^2}-\frac{1}{\bar{c}^2}\frac{\partial^2\phi}{\partial t^2}=\frac{1}{\lambda_J^2}\sin\phi \ , \qquad (1)$$

where ϕ is the phase difference of the superconducting order parameter across the junction. An approximate periodic solution to this equation for a junction of length L is obtained by summing the solutions for an infinite junction, i.e.,

TABLE I. Junction parameters.

$a=10$ Å, $L=200$ μm, $W=700$ Å, $J_c=400$ A/cm^2, $\lambda_J=20$ μm, $d=1780$ Å, λ_L(Pb-In-Au) $=1370$ Å, λ_L(Nb) $=390$ Å

$$\phi=4\sum_{n=-\infty}^{\infty}\left\{\tan^{-1}\left[\exp\left(+\frac{x-2nL-u^{+}t-L/2}{\lambda(u^{+})}\right)\right]+\tan^{-1}\left[\exp\left(-\frac{x+2nL+u^{-}t+L/2}{\lambda(u^{-})}\right)\right]\right\} \ , \qquad (2)$$

where $\lambda(u)=\lambda_J(1-u^2/\bar{c}^2)^{1/2}$ and where the $u^{\pm}$ are the velocities of the soliton and the antisoliton. The presence of a magnetic field not only modifies the boundary conditions[9] but also, and more importantly, changes the kinetic energy K by an amount $\Delta K \propto \pm H_{\text{ext}}$. As a result the velocity of the soliton u^{+} and antisoliton u^{-} are different. It is this which introduces the important field dependence to our theory.

The magnetic field *associated with the solitons* is obtained by differentiation with respect to distance, i.e., $B_s=(\hbar/2ed)\partial\phi/\partial x$. The result for B_s is straightforward to obtain but rather complicated. Each soliton is associated with a flux quantum Φ_0 corresponding to an area of dimension λ_J along the junction d and in the perpendicular direction. If we assume $L \gg \lambda_J$ then the spatial structure will be unimportant and the result will be of the form

$$B_s(x,t)=\frac{\Phi_0}{d}\sum_m[\delta(x-2mL-u^{+}t-L/2) - \delta(x-2mL-u^{-}t-L/2)] \ , \qquad (3)$$

where we have used δ functions to represent the solitons. It is important to note that this expression for B_s is odd, i.e., changes sign when $x \to -x$.

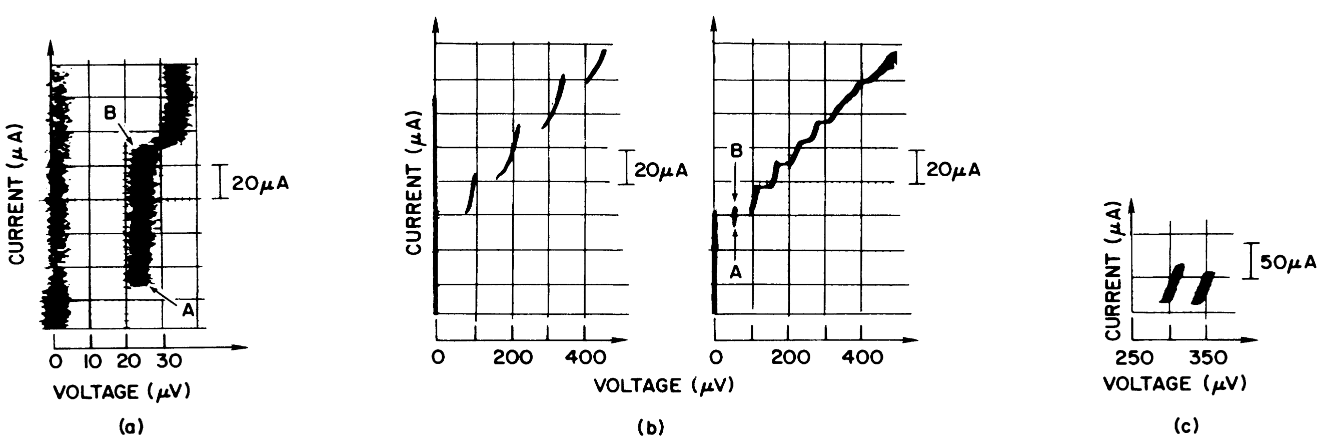

FIG. 2. (a) Shown is the steplike feature lying between A and B which we identify as the $n=2$ 24-μV ESR signal. (b) The curve to the left is for zero and to the right for finite field. The first vertical step also labeled A-B on the right and which is only present for finite fields, is identified as the $n=1$, 47-μV signal. The other branches which appear with the field might be associated with the existence of a magnetic soliton. (c) A pair of branches which differ by a shift of ~47 μV on the voltage axis.

If the susceptibility of interest were not resonant, its effect could be included in the definitions of $\bar{c}$, λ_J, and the effective damping constant. Because the susceptibility of the magnetic system *is* resonant the change $\phi \to \phi + \Phi$ caused by the magnetic system involves only relatively low frequencies and wavelengths of the order of the junction length. It is, therefore, important in calculating Φ to correctly account for the boundary conditions $[\partial\phi/\partial x = (2ed/\hbar)B_{\text{ext}}]$ at the ends of the junction. In the usual way, we accomplish this by expanding Φ or the magnetic perturbation B in terms of Fiske modes. In complex notation, the expansion for the field is

$$B = e^{i\omega t}\left(\sum_{n=\text{even}} B_n \sin(k_n x) + \sum_{n=odd} B_n \cos(k_n x)\right); k_n = \frac{n\pi}{L} . \tag{4}$$

The magnetic response is only important when ω is near that of the ESR resonant frequency. Using this, Eq. (1) can be rewritten to read

$$[1+\chi(\omega)]\frac{\partial^2\phi}{\partial x^2} - \frac{1}{\bar{c}^2}\frac{\partial^2\phi}{\partial t^2} + \frac{\partial^2\Phi}{\partial x^2} - \frac{1}{\bar{c}^2}\frac{\partial^2\Phi}{\partial t^2} = \frac{1}{\lambda_J^2}\sin\phi , \tag{5}$$

where $\chi(\omega) = \chi' - i\chi''$ is the complex susceptibility associated with the ESR and where it has been assumed that Φ is sufficiently small that it can be neglected in the argument of the sine. B is then given by

$$\frac{\partial^2 B}{\partial x^2} - \frac{1}{\bar{c}^2}\frac{\partial^2 B}{\partial t^2} = -\chi(\omega)\frac{\partial^2 B_s}{\partial x^2} . \tag{6}$$

Solving this equation gives

$$B_n = \frac{2\chi(n\omega_0)k_n^2\Phi_0}{Ld}\,\frac{1}{(n^2\omega_0^2/\bar{c}^2) - k_n^2} \times \begin{cases} [n\omega_0\Delta\omega/(\Delta\omega^2 + 4n\omega_0^2)][\sin(\frac{1}{2}\Delta\omega T)/\frac{1}{2}\Delta\omega T], & n = \text{odd}, \\ 1, & n = \text{even}, \end{cases} \tag{7}$$

where $\Delta = \frac{1}{2}k_n(u^+ - u^-)$ determined by the magnetic field B_{ext} and $\omega_0 \equiv e\bar{V}/\hbar$ measures the applied voltage. The current response associated with the ESR is then calculated by equating the time derivative of the magnetic energy to the electrical power, i.e.,

$$P = I_m\bar{V} = \frac{4WLd}{\mu_0}\frac{1}{T}\int_0^T \left(B_s\frac{\partial B}{\partial t}\right)dt . \tag{8}$$

The final result for the current I_n associated with the nth Fiske mode is

$$I_n = 2n\pi S_n^2 I_c\left(\frac{\lambda_J}{L}\right)^2\left(\frac{k_n^2}{(ne\bar{V}/\hbar\bar{c})^2 - k_n^2}\right)\chi''(ne\bar{V}/\hbar) , \tag{9}$$

where I_c is the critical current and

$$S_n = \begin{cases} 1, & n = \text{even}, \\ \left(\frac{n\omega_0\Delta\omega}{\Delta\omega^2 + 4n\omega_0^2}\right)^2\left[\left(\sin\frac{\frac{1}{2}\Delta\omega T}{\frac{1}{2}\Delta\omega T}\right)\right]^2, & n = \text{odd} . \end{cases} \tag{10}$$

The results, Eqs. (9) and (10), have several interesting properties. First the coupling of the solitons to the ESR is strong. If the junction is not too long so that λ_J is sufficiently smaller than L that the solitons are well-defined excitations but still of the same order as L then I_n is of order $I_c\chi''$ which, since χ'' is of order unity on resonance, gives a large signal of the order of the critical current. The "fundamental" signal only occurs for finite fields. It is associated with the $n=1$ Fiske mode and occurs when $\hbar\omega_r = e\bar{V}$ where ω_r is the ESR resonant frequency; this corresponding here to 47 μV. The $n>1$ signals appear as subharmonics and are associated with the corresponding order Fiske mode. The even (or odd) subharmonics are of comparable magnitude, however, the theory assumes $\lambda_J k_n < 1$ and when this inequality fails the coupling will become small. This limits the number of subharmonics which might be observed. To the sum of the I_n must be added the current

$$I_\sigma = (2\sigma L^2/\pi^2\lambda_J)\{\bar{V}/[1-(\bar{V}/V_1)]^{1/2}\} ,$$

due to resistive damping of the solitons; here σ reflects the normal conductivity and $V_1 = \hbar\pi\bar{c}/eL$ is the voltage expected for the second Fiske mode.

The magnetic susceptibility

$$\chi''(\omega) = \chi_0\omega_r\delta/[(\omega_r - \omega)^2 + \delta^2] ,$$

where δ is the width for Cr^{3+} resonance, is very strongly peaked, i.e., $\delta \ll \omega_r$. It follows that, in a current-fed junction, the resonance will appear as an almost vertical step. We identify the vertical region from A to B in Fig. 2(a) at 24 μV as such an $n=2$ step in the lowest soliton branch. The lowest vertical, finite field branch at 47 μV, and also labeled A-B in Fig. 2(b), is identified with the fundamental $n=1$ step. Lower subharmonics are not observed because the lower parts of the soliton branches are not stable at the small current end.

So much is in accord with our expectations. What is surprising is that the other branches for junction of Fig. 2(b) and the doubling of the branches seen in Fig. 2(c) imply the existence of what might be called a "magnetic soliton." We speculate that the higher voltage branches shown in Fig. 2(b) and which appear with a finite field are associated with this magnetic soliton. These branches are certainly not vertical as would be the case if they were a harmonic of the ESR signal. In fact they coincide with a part of a zero-field soliton branch moved down the voltage axis by 47 μV. The paired branches shown in Fig. 2(c) would appear to be the same branch shifted by 47 μV. These observations are consistent with the existence of a magnetic soliton which has its velocity "locked" to that associated with the ESR resonant frequency. The extra branches are then explained as being due to the addition of one slow-moving magnetic soliton which coexists and passes through the faster-moving regular solitons. The magnetic soliton sits at some point on its vertical branch and simply adds 47 μV and a small current to the branch which would exist without this soliton.

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