# Nonlinear current response of a *d*-wave superfluid

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Despite several efforts the nonlinear Meissner effect in *d*-wave superconductors, as has been discussed by Yip and Sauls in 1992, has not been verified experimentally in high-$T_c$ superconductors at present. Here, we reinvestigate the nonlinear response expected in a *d*-wave superconductor. While the linear $|\vec{H}|$ field dependence of the penetration depth, predicted by Yip and Sauls, is restricted by the lower critical field and can be masked by nonlocal effects, we argue that the upturn of the nonlinear coefficient of the *quadratic* field dependence is more stable and remains observable over a broader range of parameters. We investigate this by studying the influence of nonmagnetic impurities on the nonlinear response. We discuss the difficulties of observing this intrinsic *d*-wave signature in present day high-$T_c$ films and single crystals.
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## I. INTRODUCTION

In a $d_{x^2-y^2}$-wave superconductor, quasiparticles near the nodes of the gap give rise to an intrinsic nonlinear electrodynamic response. Yip and Sauls[1] discussed this and suggested that at sufficiently low temperatures this nonlinearity would lead to an increase in the penetration depth $\lambda$ which would vary as the magnitude of the magnetic field $|\vec{H}|$ with a coefficient that depended upon the orientation of the field relative to the nodes. Although there have been various experimental studies[2–5] of the magnetic field dependence of the penetration depth, there are at present no observations which are in agreement with the size, field, and temperature dependence predicted in Ref. 1. It is known that impurities[6] can wash out the linear $\vec{H}$ dependence, leading to an $H^2$ dependence for $\lambda$ and recently Li *et al.*[7] have argued that nonlocal effects, except for special orientations of the field, will lead to a quadratic dependence below a cross-over field of order $H_{c1}$. Thus, the magnitude $|\vec{H}|$ signature of a $d_{x^2-y^2}$-wave state in the penetration depth may be difficult at best to observe.

However, as discussed by Xu, Yip, and Sauls,[6] at higher temperatures the temperature dependence of the coefficient of the quadratic field term in the penetration depth shows a clear deviation from the exponentially decaying temperature behavior which would be observed in a fully gapped *s*-wave superconductor. In particular for a $d_{x^2-y^2}$ gap, this coefficient exhibits a $T^{-1}$ temperature dependence so that contrary to the *s*-wave case, the strength of the $H^2$ contribution increases as $T$ is decreased until it saturates due to impurities, nonlocal effects, or possibly, for the magnetic field oriented along the nodes, the crossover to the Yip-Sauls $|\vec{H}|$ dependence. It is this aspect of the nonlinear response that we wish to explore.

As we have previously discussed,[8] the quadratic term in the penetration depth leads to a nonlinear inductance which gives rise to third order intermodulation effects. This nonlinearity in fact represents a problem for superconducting communication filters and care must be taken to reduce its presence. However, from the point of view of studying the nonlinear superconducting response, microwave intermodulation effects provide a sensitive probe.[8,9] Here we present results which illustrate the type of nonlinear dependence that can be expected.

In Sec. II we study the field and temperature dependence of the nonlinear superfluid density for a clean *d*-wave superconductor. The crossover between the $|\vec{H}|$ linear Yip-Sauls regime and the quadratic regime with its divergent coefficient are discussed. Section III illustrates the corresponding behavior expected to be seen in harmonic generation and intermodulation. In Sec. IV we study the influence of nonmagnetic impurities in order to see how stable these signatures of the *d*-wave state are. We will see that the increase of the nonlinear coefficient at low temperatures appears to be more stable than the $|\vec{H}|$ linear behavior due to its restriction by the lower critical field $H_{c1}$. We discuss the difficulties in observing these signatures in present day high-$T_c$ systems. Section V contains our conclusions.

## II. THE NONLINEAR SUPERFLUID DENSITY

The current density in a superconductor is given by the sum of the superfluid flow $j_s$ and the quasiparticle backflow contribution

$$j = j_s + j_{\mathrm{qp}} \tag{1}$$

with[8]

$$j_{\mathrm{qp}} = \frac{4en}{m v_F^2} \int_0^\infty d\epsilon \times \int \frac{d\Theta}{2\pi} v_F \cos\Theta \cdot f\left( \sqrt{\epsilon^2 + \Delta^2(\Theta)} + \frac{m v_F}{ne} j_s \cos\Theta \right). \tag{2}$$

 

Here $f(\epsilon)=1/[1+\exp(\epsilon/T)]$ is the Fermi function and we have taken a simple circular Fermi surface with a $d_{x^2-y^2}$ gap. As has been discussed by Li *et al.*[7] nonlocal effects might become important for current flow along the antinodal direction for a magnetic field oriented perpendicular to the $CuO_2$ planes. However, nonlocal effects are negligible for current flow along a nodal direction. Here and in the following we will therefore study this second case using the $d$-wave angular dependence

$$\Delta(\Theta)=\Delta(T)\sin 2\Theta. \tag{3}$$

Here, for convenience, we have chosen our coordinates such that $\Theta=0$ corresponds to the nodal direction. We have checked that qualitatively very similiar results are found for current flow along the antinodal direction, if nonlocal effects are neglected. The superfluid density $n_s$ is defined by

$$j=\frac{n_s}{n}j_s \tag{4}$$

so that

$$\frac{n_s}{n}=1+4\frac{j_c}{j_s}\int_0^\infty\frac{d\epsilon}{\Delta_0}\times\int\frac{d\Theta}{2\pi}\cos\Theta\cdot f\left(\sqrt{\epsilon^2+\Delta^2(\Theta)}+\Delta_0\frac{j_s}{j_c}\cos\Theta\right) \tag{5}$$

with $j_c=ne\Delta_0/mv_F$ the pair breaking current density. In the limit $j_s\to 0$, we have

$$\frac{n_s(j_s=0)}{n}=1+4\int_0^\infty d\epsilon\times\int\frac{d\Theta}{2\pi}\cos^2\Theta\cdot\frac{df}{d\epsilon}[\sqrt{\epsilon^2+\Delta^2(\Theta)}]. \tag{6}$$

Setting

$$\delta n_s=n_s(j_s=0)-n_s(j_s) \tag{7}$$

we have for the nonlinear contribution to the superfluid density

$$\frac{\delta n_s(j_s)}{n}=-4\frac{j_c}{j_s}\int_0^\infty\frac{d\epsilon}{\Delta_0}\times\int\frac{d\Theta}{2\pi}\cos\Theta f\left(\sqrt{\epsilon^2+\Delta^2(\Theta)}+\Delta_0\frac{j_s}{j_c}\cos\Theta\right)+4T\int_0^\infty\frac{d\epsilon}{\Delta_0}\int\frac{d\Theta}{2\pi}\cos^2\Theta\frac{df}{d\epsilon}[\sqrt{\epsilon^2+\Delta^2(\Theta)}]. \tag{8}$$

At small current flow this is related to the nonlinear change in penetration depth $\Delta\lambda$ via

$$\frac{\Delta\lambda}{\lambda}\simeq\frac{1}{2}\frac{\delta n_s}{n}. \tag{9}$$

It is this nonlinear part of the superfluid density that we will study.

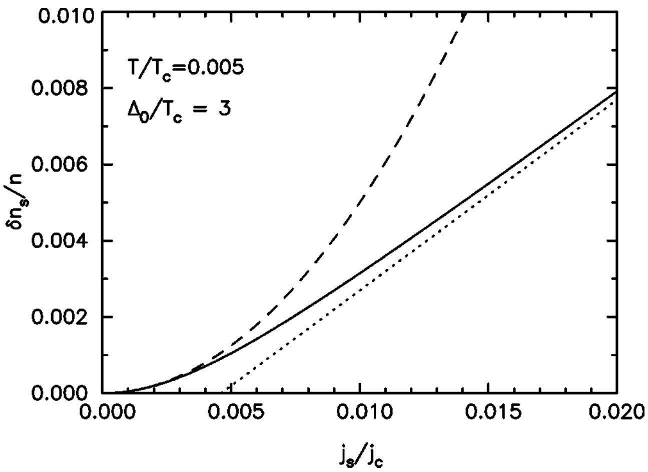


FIG. 1. The nonlinear part of the superfluid density $\delta n_s/n$ as a function of superfluid current density $j_s/j_c$ for temperature $T/T_c=0.005$ (solid line). The dashed line shows the parabolic low-$j_s$ expansion Eq. (10) and the dotted line the Yip-Sauls result Eq. (14).

There are some simple limiting cases. As discussed by Yip and Sauls, when $j_s/j_c\ll T/\Delta_0$, then one can expand Eq. (8) in powers of $j_s/j_c$ so that

$$\frac{\delta n_s}{n}=\beta(T)\left(\frac{j_s}{j_c}\right)^2 \tag{10}$$

with

$$\beta(T)=-\frac{2}{3}\Delta_0^2\int_0^\infty d\epsilon\int\frac{d\Theta}{2\pi}\cos^4\Theta\cdot\frac{d^3f}{d\epsilon^3}[\sqrt{\epsilon^2+\Delta^2(\Theta)}]. \tag{11}$$

At low temperatures this expression yields

$$\beta(T)\simeq\frac{1}{12}\frac{\Delta_0}{T}, \tag{12}$$

the $1/T$ divergence mentioned above. Alternatively, in the low temperature limit where

$$\frac{n_s(j_s=0)}{n}=1-2\ln 2\left(\frac{T}{\Delta_0}\right) \tag{13}$$

and $j_s/j_c\gg T/\Delta_0$ we have the Yip-Sauls result[1]

$$\frac{\delta n_s(j_s)}{n}=\frac{1}{2}\frac{|j_s|}{j_c}-2\ln 2\left(\frac{T}{\Delta_0}\right). \tag{14}$$

For the calculations presented in the following we choose $\Delta_0/T_c=3$, a value that fits low temperature penetration depth data on YBCO.[10]

For $T/T_c=0.005$ the two limits in Eqs. (10) and (14) are shown in Fig. 1 along with the numerical result obtained by numerically integrating Eq. (8). Here we see a crossover from a quadratic dependence on $j_s/j_c$ for $j_s/j_c<T/T_c$ to a linear dependence when $j_s/j_c$ becomes larger than $T/T_c$. Figure 2 shows a sequence of curves for $\delta n_s/n$ versus $j_s/j_c$ for different reduced temperatures. At larger values of $j_s/j_c$, $\delta n_s/n$ has approximately the same slope, but the curves are shifted down by an amount proportional to $T/T_c$. As has been pointed out by Yip and Sauls[1] and Li *et al.*[7] this linear $j_s/j_c$ dependence can at best only be observable at low temperatures due to the fact that a type II superconductor will enter the vortex state, if the current density level $j_s/j_c$

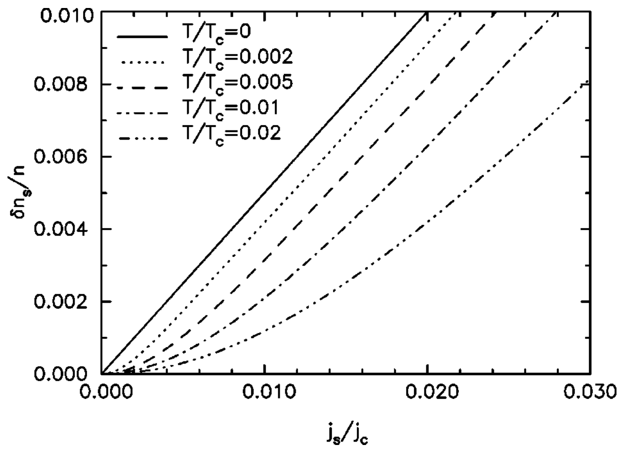


FIG. 2. The nonlinear part of the superfluid density $\delta n_s/n$ as a function of superfluid current density $j_s/j_c$ for different reduced temperatures $T/T_c$.

reaches the lower critical field $H_{c1}/H_c \simeq 0.01$ for high-$T_c$ superconductors. At small values $j_s/j_c < T/T_c$, $\delta n_s/n$ enters the parabolic regime and the curvature $\beta(T)$ given in Eq. (11) diverges as $1/T$ upon lowering the temperature.[6,8] At the same time the convergence radius of the Taylor expansion decreases like $T/T_c$, showing the crossover to the nonanalytic Yip-Sauls result at $T=0$.

## III. HARMONIC GENERATION AND INTERMODULATION

The nonlinear response to an applied microwave field such that the microwave frequency is small compared to the quasiparticle relaxation time can be determined from Eqs. (8) and (4). If

$$j_s(t) = j_{s0} \sin \omega t \tag{15}$$

then

$$j(t) = \frac{n_s[j_s(t)]}{n} j_s(t). \tag{16}$$

Now, only odd frequency terms arise since $n_s(j_s) = n_s(-j_s)$. For example, the third harmonic $j_3 \sin 3\omega t$ has an amplitude

$$j_3 = \frac{1}{\pi} \int_0^{2\pi} dx \sin 3x \frac{n_s(j_{s0} \sin x)}{n} j_{s0} \sin x. \tag{17}$$

In the "high temperature limit," $T/T_c \gg j_s/j_c$ we find using Eq. (10)

$$j_3 = \frac{1}{4} \beta(T) \frac{j_{s0}^3}{j_c^2} \tag{18}$$

and in the "low temperature limit," $T/T_c \ll j_s/j_c$ using Eq. (14) we have

$$j_3 = \frac{4}{15\pi} \frac{j_{s0}^2}{j_c}. \tag{19}$$

Figure 3 shows a double logarithmic plot of $j_3/j_c$ versus $j_{s0}/j_c$ for three different values of $T/T_c$ and the crossover from Eq. (18) (solid line) to Eq. (19) (dotted line) is clearly

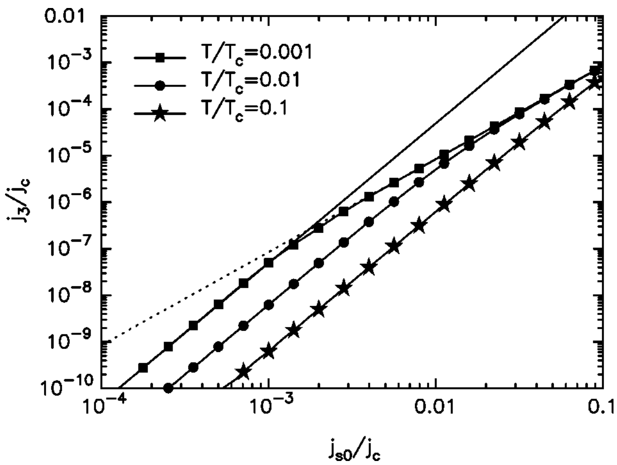


FIG. 3. The amplitude $j_3/j_c$ of the third harmonic generated by the nonlinear superfluid density as a function of the amplitude of the fundamental current density $j_{s0}/j_c$ for different reduced temperatures $T/T_c$. The two limiting cases Eqs. (18) (for $T/T_c = 0.001$) and (19) are shown as the solid and the dotted line, respectively.

seen.[9] Note, that due to the prefactor $\beta(T)$ in Eq. (18) the nonlinear response $j_3/j_c$ in the low $j_{s0}/j_c$ regime increases, when the temperature is lowered. Figure 4 shows the temperature dependence of $j_3/j_c$ for $j_{s0}/j_c = 0.01$ along with the high temperature limit from Eq. (18). The third harmonic amplitude $j_3$ follows the $1/T$ divergence until $T/T_c$ falls below $j_{s0}/j_c = 0.01$. Below that point $j_3$ saturates due to the crossover to the Yip-Sauls limit Eq. (19). This low temperature peak in the third harmonic amplitude is a consequence of the nodes of the $d$-wave state and does not exist for an $s$-wave superconductor.[8]

One can also explore two-tone intermodulation which is important in communication filter applications. Here

$$j_s(t) = j_{s1} \sin \omega_1 t + j_{s2} \sin \omega_2 t \tag{20}$$

with $\omega_1$ and $\omega_2$ close in frequency so that the intermodulation frequency $2\omega_1 - \omega_2$ lies within the pass band of the filter. In this case we find for the amplitude $j_{2\omega_1-\omega_2}$ of the intermodulation response at high temperatures[11]

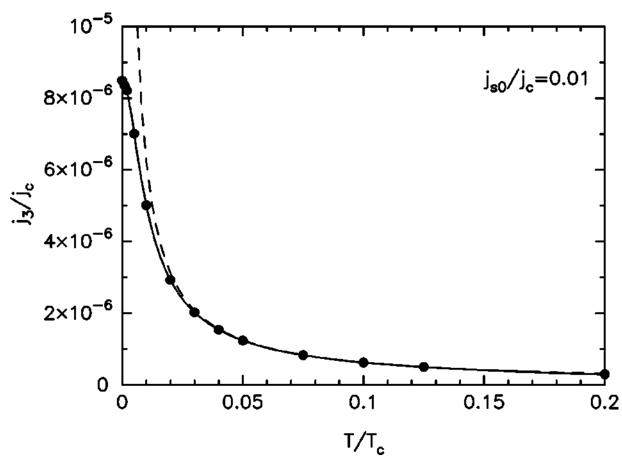


FIG. 4. Temperature dependence of $j_3/j_c$ for $j_{s0}/j_c = 0.01$ (solid line). The dashed line shows the high temperature limit Eq. (18). Below $T/T_c = j_{s0}/j_c = 0.01$, the third harmonic amplitude $j_3/j_c$ saturates due to the crossover to the Yip-Sauls regime Eq. (19).

$$j_{2\omega_1-\omega_2}=\frac{3}{4}\beta(T)\frac{j_{s1}^2 j_{s2}}{j_c^2} \tag{21}$$

while at low temperatures, when $j_{s2}\ll j_{s1}$

$$j_{2\omega_1-\omega_2}=\frac{1}{2\pi}\frac{j_{s2}^2}{j_c}, \tag{22}$$

and when $j_{s1}\ll j_{s2}$

$$j_{2\omega_1-\omega_2}=\frac{2}{3\pi}\frac{j_{s1}j_{s2}}{j_c}. \tag{23}$$

Thus, at fixed $j_{s1}$ and $j_{s2}$ the temperature dependence of $j_{2\omega_1-\omega_2}$ will have the same qualitative behavior as $j_3(T)$ in Fig. 4.

This analysis shows that measurements of third harmonics and intermodulation allow us to directly access the temperature dependence of the nonlinear coefficient $\beta(T)$, utilizing Eqs. (18) or (21). In principle, this allows to probe experimentally whether this low temperature peak in $\beta$ exists or not.

## IV. INFLUENCE OF IMPURITIES

Now we wish to consider the influence of nonmagnetic impurities on the nonlinear response discussed in the previous sections in order to see how stable the $d$-wave signatures in the nonlinear response are. In the presence of impurities the total current density is given by[6]

$$j=-2j_c\int_{-\pi/2}^{\pi/2}\frac{d\Theta}{2\pi}\cos\Theta\int_{-\infty}^{\infty}\frac{d\omega}{\Delta_0}f(\omega)\cdot[N_+(\Theta,\omega)-N_-(\Theta,\omega)], \tag{24}$$

where $N_\pm(\Theta,\omega)$ is the density of states for the comoving and countermoving quasiparticles, respectively:

$$N_\pm(\Theta,\omega)=\mathrm{Im}\frac{\tilde{\omega}\pm\Delta_0(j_s/j_c)\cos\Theta}{\sqrt{\Delta^2(\Theta)-[\tilde{\omega}\pm\Delta_0(j_s/j_c)\cos\Theta]^2}}. \tag{25}$$

Here, $\tilde{\omega}(\omega)$ is the renormalized frequency and has to be determined by the selfconsistent equations[12,13]

$$g_0=-\int_{-\pi}^{\pi}\frac{d\Theta}{2\pi}\frac{\tilde{\omega}+\Delta_0(j_s/j_c)\cos\Theta}{\sqrt{\Delta^2(\Theta)-[\tilde{\omega}+\Delta_0(j_s/j_c)\cos\Theta]^2}}, \tag{26}$$

$$\tilde{\omega}=\omega-\Gamma\frac{g_0}{c^2-g_0^2}. \tag{27}$$

Here, $c$ is the cotangent of the scattering phase shift and $\Gamma$ the scattering rate. Using Eqs. (24)–(27) along with Eqs. (4) and (7) we can extract $\delta n_s(j_s)/n$ in the presence of nonmagnetic impurities.

In Fig. 5 we show $\delta n_s(j_s)/n$ in the unitarity limit $c=0$ with $\Gamma=0.003T_c$, a typical value in a range that has been used to fit low temperature penetration depth[10] and thermal conductivity data.[14] Due to the finite impurity scattering $\delta n_s/n$ now stays quadratic at low $j_s/j_c$ down to $T=0$. Now, the linear $j_s/j_c$ regime is only entered at higher values of $j_s/j_c\gg\Gamma/T_c$. The restriction due to the lower critical field $j_s/j_c\leqslant H_{c1}/H_c\simeq 0.01$ can make the linear $j_s/j_c$ regime unobservable in this case, as can nonlocal effects for current flow along the antinodal direction.[7]

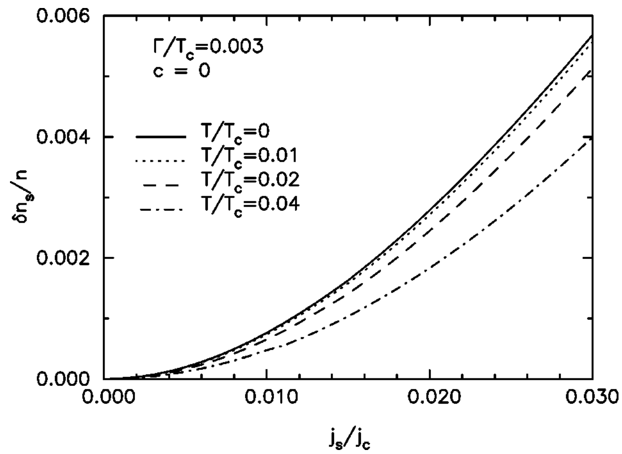


FIG. 5. $\delta n_s/n$ as a function of $j_s/j_c$ for different reduced temperatures $T/T_c$ in the presence of nonmagnetic impurities in the unitarity limit $c=0$. The scattering rate is $\Gamma=0.003T_c$.

In order to find the nonlinear coefficient $\beta(T/T_c)$ it is advantageous to perform the calculation on the imaginary frequency axis. Then we find

$$\beta(T)=2\Delta_0^2\int_{-\pi}^{\pi}\frac{d\Theta}{2\pi}\cos^4\Theta\times\pi T\sum_{\omega_n>0}\Delta^2(\Theta)\mathrm{Re}\left\{\frac{4\tilde{\omega}_n^2-\Delta^2(\Theta)}{[\tilde{\omega}_n^2+\Delta^2(\Theta)]^{7/2}}\right\}. \tag{28}$$

Here, $\tilde{\omega}_n$ are the renormalized Matsubara frequencies which have to be determined self-consistently by the imaginary axis counterparts of Eqs. (26) and (27):

$$\tilde{\omega}_n=\omega_n+\Gamma\frac{\overline{g}_0}{c^2+\overline{g}_0^2}, \tag{29}$$

$$\overline{g}_0=\int_{-\pi}^{\pi}\frac{d\Theta}{2\pi}\frac{\tilde{\omega}_n}{\sqrt{\Delta^2(\Theta)+\tilde{\omega}_n^2}} \tag{30}$$

with $\omega_n=(2n+1)\pi T$ being the unrenormalized Matsubara frequencies.

Figure 6 shows $\beta(T/T_c)$ for different scattering rates. Now, the $1/T$ divergence is cut off at low temperatures by the impurity scattering. However, a peak, signifying the underlying $d$-wave nature of the superconducting gap, still remains unless $\Gamma/T_c$ becomes of the order of 0.1. Qualitatively similar results are found for nonunitary scattering $c\neq 0$. The influence of impurities becomes even less pronounced, though.

As an illustration we show the density of states $N(\omega)$ in the presence of impurities in Fig. 7. Here one can see the impurity states generated at low energies $\omega/\Delta_0<0.05$.[12,15]

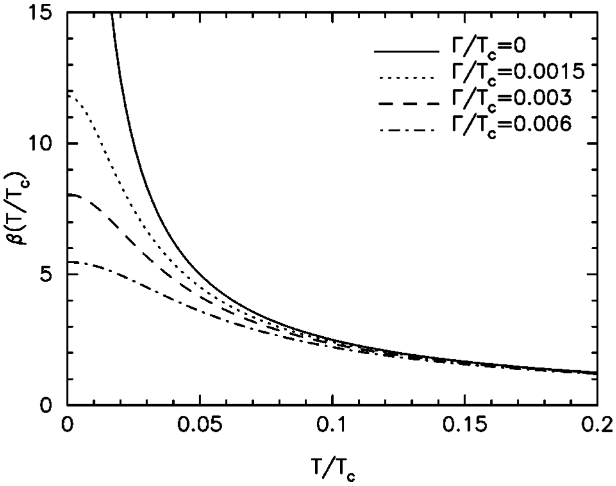


FIG. 6. Temperature dependence of the nonlinear coefficient $\beta(T/T_c)$ for different scattering rates $\Gamma$ in the unitarity limit.

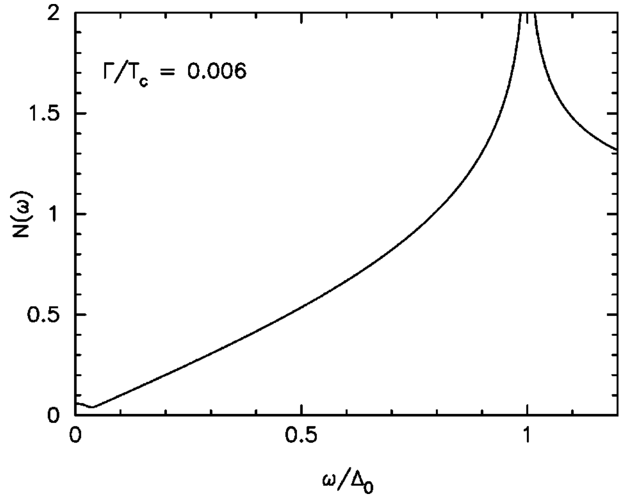


FIG. 7. The density of states $N(\omega)$ in the presence of impurities in the unitarity limit for $\Gamma=0.006T_c$. Close to the Fermi level $\omega/\Delta_0<0.05$ impurity states are generated. However, over a broad region $N(\omega)$ still varies linearly with $\omega$ as in the clean $d$-wave state.

However, at higher energies there is still a broad region where $N(\omega)$ varies linearly with $\omega$. At not too low tempera tures the nonlinear response still picks up this linear variation resulting in an upturn of the nonlinear coefficient $\beta(T/T_c)$ upon lowering the temperature.

This signature of the $d$-wave state should remain observable over a much broader range of parameters than the $|\vec{H}|$-linear regime discussed by Yip and Sauls. While the $|\vec{H}|$-linear regime is limited by the lower critical field $\Gamma/T_c \ll H_{c1}/H_c \simeq 0.01$, this peak in $\beta(T/T_c)$ will remain until $\Gamma/T_c$ becomes of the order of 0.1, making it a better candidate for a search of $d$-wave behavior in the nonlinear response. From the study in Ref. 7 we expect that this upturn of $\beta$ should also remain visible, if one includes nonlocal effects for current flow along the antinodal direction.

Some remarks concerning the difficulties of observing this behavior should be made at this point. In order to extract the coefficient $\beta$ from measurements of the $\vec{H}$-field dependence of the penetration depth $\lambda$ one would need a very high resolution, as can be estimated from Eqs. (9) and (10): even if $j_s/j_c \simeq 0.01$ and for $\beta$ we take a typical value of 4, we find $\Delta\lambda/\lambda \simeq 2\times10^{-4}$ which challenges existing techniques.[4] A more direct way to measure $\beta$ would be harmonic generation or intermodulation utilizing Eqs. (18) and (21) as has been discussed in the previous section. A study of the temperature dependence of intermodulation in high-$T_c$ (TBCCO) films has been done in Ref. 16. In that study no increase in intermodulation was found down to the lowest temperatures of 25 K, which might not be small enough, however. A similar result was found in measurements of third harmonic generation in YBCO films.[17] These studies also showed that the absolute magnitude of the nonlinear response is higher than expected from the intrinsic $d$-wave response discussed here. This and the study in Ref. 4 indicates that other sources of nonlinear behavior, as for example weak link grain boundaries which act as Josephson Junctions,[18] might dominate the nonlinear response in present day high-$T_c$ films. Thus, the difficulties of observing $d$-wave behavior in the nonlinear response are mainly related to the existence of extrinsic effects even in the best presently available systems. Nevertheless, we suggest that a temperature dependent measurement of harmonic generation or intermodulation in the highest quality single crystals available today provides the best hope of observing $d$-wave behavior in the nonlinear response.

## V. CONCLUSIONS

We studied the temperature and field dependence of the nonlinear electrodynamic response in a $d$-wave superconductor. The signatures of the $d$-wave state are the $|\vec{H}|$-linear regime, as discussed by Yip and Sauls, and an upturn of the nonlinear coefficient $\beta$ in the quadratic regime at low temperatures. This coefficient can be directly measured by harmonic generation and intermodulation. While the $|\vec{H}|$-linear regime is limited by the lower critical field and can be masked by impurity scattering and nonlocal effects, the upturn of the coefficient $\beta$ appears to be more stable and should remain observable over a broader range of parameters. We showed this explicitly by studying the influence of nonmagnetic impurity scattering. It is possible, however, that in present day high-$T_c$ films and single crystals extrinsic effects still dominate the nonlinear response, masking this intrinsic signature of the $d$-wave symmetry.

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