Theory of far-infrared absorption in superconducting composites

J. Garner* and D. Stroud
Department of Physics, The Ohio State University, Columbus, Ohio 43210
(Received 15 April 1983)

A simple model is described for the frequency-dependent conductivity of normal-superconducting composites. The superconducting component is characterized by the Mattis-Bardeen conductivity, and the composite conductivity is determined via the effective-medium approximation. Near the percolation threshold p_c there is strong absorption below twice the superconducting gap. Scaling arguments indicate that our results have validity beyond mean-field theory. The model is in qualitative agreement with recent experiments on granular NbN films.

I. INTRODUCTION

Composite superconductors behave very differently from ordinary, bulk superconductors.[1,2] For example, their resistive transition is broadened over a large fraction of a kelvin instead of being abrupt and nearly discontinuous,[3–8] their specific-heat anomaly at the transition is of a different shape than in bulk superconductors,[9,10] and they have upper critical fields which depend on temperature in a unique way.[11] Numerous models have been proposed to explain various aspects of this behavior. They draw on percolation theory,[12–15] and analogies with the classical two-component spin models,[16–18] especially (in quasi-two-dimensional films) with the vortex-unbinding transition originally proposed by Kosterlitz and Thouless and Berezinskii for superfluid ^{4}He films.[19–23] The connection between these phenomena is fascinating, and still being unraveled.

In this paper we present a simple model for the far-infrared properties of composite superconductors that leads to a conspicuous absorption below twice the superconducting energy gap Δ, or equivalently, a substantial real part in the effective frequency-dependent conductivity of the composite below this energy. The size of this real part is predicted to be greatest near the percolation threshold, that is, the volume fraction p_c of superconductor at which it first forms a connected cluster extending throughout the sample. We calculate the effective conductivity of a normal-superconducting (N-S) composite using the effective-medium approximation[24,25] (EMA) and find results in excellent qualitative agreement with recent experiments in granular NbN films.[26] The general features of our results do not depend on the EMA and we present arguments why they should still persist in a more exact treatment of percolation, showing that our predictions are consistent with an earlier scaling treatment of the percolation transition in the finite-frequency regime.

II. FORMALISM AND RESULTS

A. Dilute limit

The essence of the enhanced absorption can be understood very simply. We consider a superconductor with a single inclusion of normal metal. The complex frequency-dependent conductivity of the superconducting host, $\sigma_s(\omega)$, may be modeled by the well-known Mattis-Bardeen[27] form,

$$\sigma_s(\omega)=\frac{iG_0}{\omega}+\sigma_s'(\omega)\ , \tag{1}$$

where at temperature $T=0$, $G_0=\sigma_n'(\pi\Delta/\hbar)$.[28] Here σ_n' is the frequency-independent and real conductivity of the superconductor in its normal state, and $\mathrm{Re}\sigma_s'(\omega)$ vanishes except above $2\Delta/\hbar$. We take σ_n, the conductivity of the normal component, to be independent of frequency in the range of interest and real. If an electric field $\vec{E}$ is applied to the superconductor, then the field $\vec{E}_{\rm in}$ within the ellipsoidal normal inclusion is uniform (neglecting displacement currents; this neglect is discussed below) and given by

$$\vec{E}_{\rm in}=\vec{E}_{\rm out}[(g+1)\sigma_s]/(\sigma_n+g\sigma_s)\ ,$$

where g is related to the effective depolarization factor for the ellipsoid ($g=2$ for spheres, $g=1$ for highly prolate ellipsoids with field perpendicular to the long axis). Since $\vec{E}_{\rm in}$ is nonzero, and since the current within the normal inclusion is *in* phase with the field, there will be dissipation and hence a real component to the effective conductivity $\sigma^e(\omega)$ of the composite as a whole, even for frequencies below $2\Delta/\hbar$. The magnitude of the dissipation is $\mathrm{Re}(\vec{J}\cdot\vec{E}^*/2)$ integrated over the volume of the composite, $\vec{J}$ being the current density.

If we only include the first (London) term in Eq. (1), (i.e., we assume frequencies below the gap), this dissipation gives for the real part of the effective conductivity,

$$\mathrm{Re}\sigma_e(\omega)\equiv\sigma_1^e(\omega)=\frac{(1-p)(g+1)^2\sigma_n G_0^2}{\omega^2\sigma_n^2+g^2G_0^2}\ , \tag{2}$$

where p is the volume fraction of superconductor. In the limit of very low frequency, this leads to a dissipation proportional to σ_n, the conductivity of the normal metal, while at other frequencies, the dissipation has a Lorentzian shape centered at zero frequency and of halfwidth proportional to G_0/σ_n. This dissipation is somewhat similar to the excitation of quasiparticles in bulk superconductors at finite temperatures, which also leads to below-gap absorption. In the present case, however, the below-gap absorption is due solely to the excitation of electrons in the normal-metal inclusion within the superconductor.

B. Effective-medium approximation

At larger concentrations of normal metal, the effective conductivity at finite frequencies cannot be calculated exactly. We estimate it using the EMA, according to which the fields and currents within each grain of the composite are calculated as if the grain were an ellipsoid embedded in a medium with a self-consistently-determined effective complex conductivity $\sigma_e(\omega)=\sigma_1^e+i\sigma_2^e$.[29] In order to include the effects of displacement currents, we work in terms of dielectric constants. The complex dielectric function $\epsilon_e(\omega)$ satisfies the equation

$$p\frac{\epsilon_s(\omega)-\epsilon_e(\omega)}{\epsilon_s(\omega)+g\epsilon_e(\omega)}+(1-p)\frac{\epsilon_n-\epsilon_e(\omega)}{\epsilon_n+g\epsilon_e(\omega)}=0\ , \qquad (3)$$

and the dielectric functions and conductivities are related by

$$\epsilon_\alpha(\omega)=1+\frac{4\pi i}{\omega}\sigma_\alpha(\omega),\quad \alpha=s,n,e\ , \qquad (4)$$

where p is as previously the volume fraction of superconductor. This simple mean-field approximation is still enough to give the essential physics. We have carried out calculations based on Eq. (3) for $g=2$ (spherical inclusions) and $g=1$ (cylinders). The first is appropriate for three-dimensional samples of compact normal and superconducting grains, and the second for thin-film superconductors in which the grains can be viewed as slices of cylinders. Representative results are shown in Fig. 1, which displays the real part of the effective conductivity

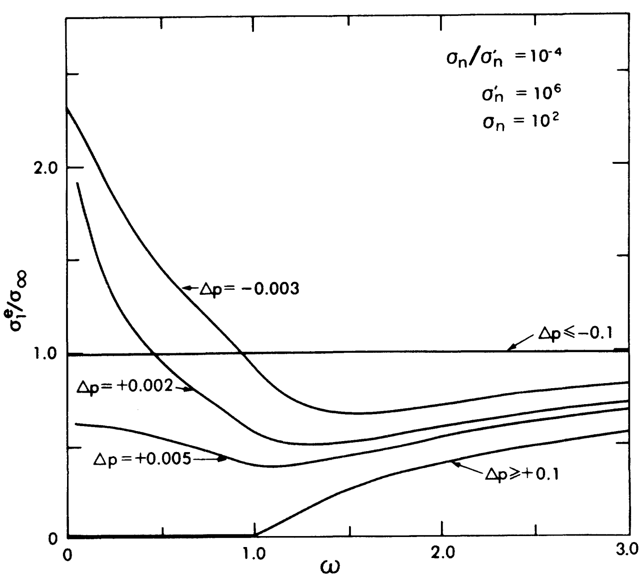

FIG. 1. Calculated real part of the effective conductivity, $\sigma_1^e(\omega)$, of a thin-film normal-superconducting composite, plotted in units of σ_∞, the dc conductivity of the composite in its normal state. The quantity σ_n/σ_n' is the ratio of the conductivity of the normal component to that of the superconducting component in its normal state. Various values of the superconducting volume fraction p are plotted. $p_c=0.5$ is the percolation threshold and $\Delta p\equiv p-p_c$. Conductivity units are such that $2\Delta/\hbar=1$.

for thin-film superconductors ($g=1$), using Eq. (3), and assuming that the ratio $\sigma_n/\sigma_n'=10^{-4}$, so that the superconducting component of the composite is a much better conductor in its normal state than is the normal component. σ_∞ is the composite dc conductivity when the superconducting component is in its normal state; it also is found via the EMA. σ_1^e/σ_∞ is plotted for several values of p, both above and below the percolation threshold p_c ($=0.5$ in the EMA for this geometry). All our results are for zero temperature. In our units (we have set $\hbar/2\Delta=1$) $\sigma_n'=10^6$, which corresponds to a normal-metal conductivity roughly that of Pb.

We distinguish four regimes of concentration:

(i) For p well above p_c, $\sigma_1^e(\omega)/\sigma_\infty$ is indistinguishable from that of a bulk superconductor. The real part has a δ function at zero frequency, corresponding to perfect conductivity, and a Mattis-Bardeen–type part for $\omega>2\Delta/\hbar$. The effective optical gap is that of the superconducting component.

(ii) For p sufficiently near p_c, but still above it, a noticeable bump in σ_1^e/σ_∞ develops below $2\Delta/\hbar$. This is superimposed on a δ function at zero frequency, whose strength diminishes as p_c is approached from above. The strength of the bump grows steadily with decreasing p, relative to the strength of the Mattis-Bardeen–type part above $2\Delta/\hbar$. The minimum of σ_1^e/σ_∞ lies very near $2\Delta/\hbar$ except for concentrations very close to p_c, where it starts to move above $2\Delta/\hbar$ and to become quite shallow.

(iii) At $p=p_c$ and for $\hbar\omega \ll 2\Delta$ we obtain from Eq. (3) above the analytic result,

$$\sigma_1^e(\omega)=-\sigma_2^e(\omega)=\left[\frac{\pi}{4}\sigma_n\sigma_n'\frac{2\Delta}{\hbar\omega}\right]^{1/2}\sim[\sigma_n \mathrm{Im}\sigma_s(\omega)]^{1/2}\ . \qquad (5)$$

(iv) For p below but near p_c, the perfect-conductivity δ function disappears, but the other features remain as in (ii).

The imaginary part, σ_2^e, of the effective conductivity follows from Eq. (3) as well. We have calculated it and find that above p_c the most important feature below the

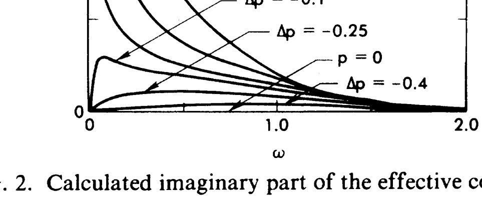

FIG. 2. Calculated imaginary part of the effective conductivity, $-\sigma_2^e(\omega)$, corresponding to the composite of Fig. 5.

gap is an inductive $1/\omega$ behavior arising from the zero-frequency δ function in the real part, but this becomes weaker near p_c (cf. Fig. 2). Experimentally, it is σ_1^e that is measured and then used to determine σ_2^e by use of the Kramers-Kronig relations.

We turn next to a comparison between our results and experiment. The results presented here bear a close resemblance to those obtained recently by Karecki *et al.* for granular NbN films.[26] Their far-infrared measurements yield values of σ_1^e/σ_∞ that show little deviation from Mattis-Bardeen behavior except for samples with resistance of 500 Ω/sq or greater. At these resistances a below-gap rise in σ_1^e/σ_∞ develops very much as shown in our Fig. 1. The resemblance seems to hold not only for the general features but also for many details: in particular, the way in which σ_1^e/σ_∞ changes with decreasing normal-state conductance, the relative importance of below-gap and above-gap conductivity, and the way in which the minimum in the frequency-dependent conductivity moves slightly higher in frequency as the normal-state conductance is lowered below the critical (presumably the percolation) value. We have not made a quantitative comparison as yet because of ambiguities in determining the value of σ_n appropriate to their experiments.

We have carried out a number of calculations similar to those shown in Fig. 1, for different conductivity ratios and for three-dimensional ($g=2$) as well as thin-film samples. In every case, the behavior shown in Fig. 1 is found. However, one additional trend is noted: As the ratio σ_n/σ_n' becomes smaller, the concentration range of conspicuous (below-gap) absorption also diminishes. When $\sigma_n/\sigma_n'=10^{-6}$, for example, (cf. Fig. 3) the below-gap absorption turns on very abruptly, and would probably be experimentally undetectable for $|p-p_c|\gtrsim 0.003$. This would correspond to onset at a very well-defined value of effective normal-state conductance of the composite, as is, in fact, seen experimentally. By contrast, if $\sigma_n/\sigma_n'=10^{-2}$ or 1 (cf. Figs. 4 and 5), below-gap absorption would be seen far above the percolation threshold and would vary much more gradually with concentration. Furthermore, these results would not change much if a more exact (i.e., non-EMA) approximation were used for the effective conductivity.

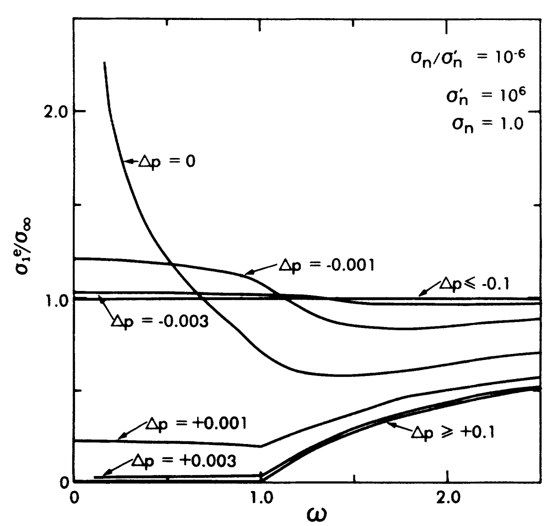

FIG. 3. Same as Fig. 1, but for $\sigma_n/\sigma_n'=10^{-6}$.

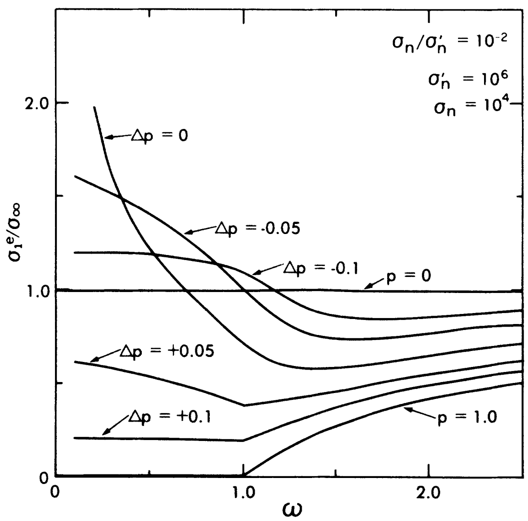

FIG. 4. Same as Fig. 1, but for $\sigma_n/\sigma_n'=10^{-2}$.

III. SCALING THEORY

The critical behavior of $\sigma_1^e(\omega)$ very near the percolation threshold deserves special discussion since it cannot be accurately treated within the mean-field EMA. A scaling description is given in Ref. 30. Application of their Eqs. (6)—(9) to the present model shows that the below-gap rise in $\sigma_1^e(\omega)/\sigma_\infty$ has a height of $\sigma_1^e(0)/\sigma_\infty$ and halfwidth $\Delta\omega$ (i.e., width at half maximum) of

$$\frac{\sigma_1^e(0)}{\sigma_\infty}=\frac{\sigma_n}{\sigma_\infty}\,|\Delta p\,|^{-s}\,, \tag{6}$$

$$\Delta\omega=\frac{\sigma_n'}{\sigma_\infty}\frac{\pi\Delta}{\hbar}\,|\Delta p\,|^{s+t}=\frac{\sigma_\infty}{\sigma_n}\frac{\pi\Delta}{\hbar}\,|\Delta p\,|^{s},\ \ \text{for } p>p_c\ , \tag{7}$$

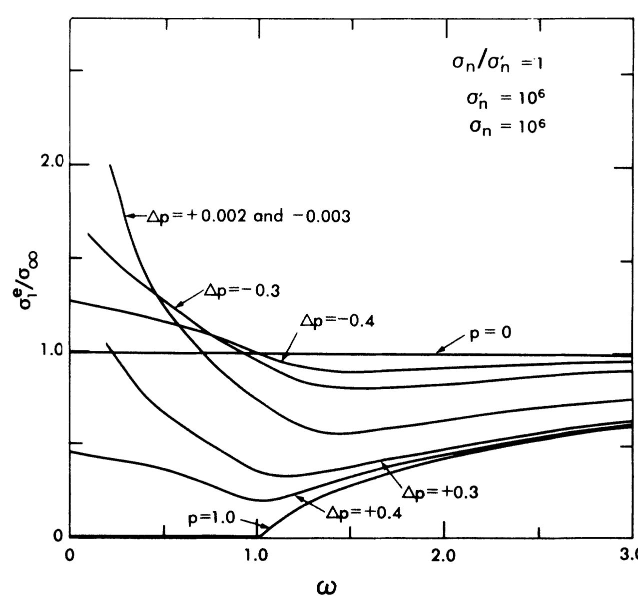

FIG. 5. Same as Fig. 1, but for $\sigma_n/\sigma_n'=1$.

$$\sigma_2^e(\omega)/\sigma_\infty = G_0(\Delta p)^t/\sigma_\infty\omega, \quad \text{for } p > p_c \;, \tag{8}$$

where $\Delta p = p - p_c$, and s and t are the usual percolation critical exponents,[12,13,31] $s = t = 1.3$ in two dimensions, $s = t = 1$ in the EMA, and σ_∞ is the dc composite conductivity in its normal state. In writing Eq. (7) we have assumed $\sigma_n' \gg \sigma_n$ and hence $\sigma_\infty = \sigma_n'(\Delta p)^t$. Equation (6) implies that $\sigma_1^e(0)/\sigma_\infty$ becomes comparable to unity when $\Delta p \sim (\sigma_n/\sigma_\infty)^{1/s}$. Thus the below-gap absorption becomes comparable to that above the gap only very close to p_c when the normal constituent is poorly conducting. This effect is already seen in the EMA. At this value of Δp, the halfwidth $\Delta\omega$ is approximately $\pi\Delta/\hbar$, comparable to the optical gap, as again predicted by the EMA. Also, as in the scaling picture of Ref. 30, the inductive part of the conductivity (i.e., the magnitude of G, the strength of the δ function) diminishes as $|\Delta p|^t$ near the threshold, and thus becomes notable at ever lower frequencies as p approaches p_c. Since σ_∞ is also proportional to $|\Delta p|^t$, we have that $G \propto \sigma_\infty \propto R_\square^{-1}$ where $R_\square$ is the resistance per square of the composite in its normal state.

Effects analogous to those predicted above are likely to occur not only in "classical" *N-S* composites but also in disordered networks of Josephson junctions and normal links, which might make a better model for some experimental systems than a macroscopic composite. In the limit of a small ac voltage drop across a single Josephson junction, the junction would have (i) an inductive conductivity, arising from zero resistance, and (ii) a "photon-assisted hopping" contribution from excitation of Cooper pairs across the junction and across the optical gap. Contribution (ii) would not contribute to the real part of the conductance except above a threshold frequency. Thus the junction would have a conductance similar *in form* to the Mattis-Bardeen conductivity, and a normal-Josephson network should exhibit a complex effective conductance similar to that predicted here.

IV. SUMMARY

We have presented a generalization of the Mattis-Bardeen conductivity to the case of *N-S* composites. The calculation leads to the prediction of a strong absorption in such composites below the optical superconducting gap, $2\Delta/\hbar$, particularly near the percolation threshold. We have calculated the zero-temperature complex conductivity of such a composite both in the dilute limit and within the EMA and found qualitative agreement with recent transmission measurements. Near the percolation threshold for superconduction the composite was found to exhibit a considerable enhancement in absorption relative to the normal state. Finally, we have obtained the scaling behavior of the complex conductivity near the percolation threshold. More detailed experimental tests of the behavior described here would certainly be useful in verifying these predictions, and, in particular, with better characterized materials composing composites of well-known volume fractions.

ACKNOWLEDGMENTS

We would like to thank Professor S. Perkowitz and Dr. G. L. Carr for valuable discussions and for describing their experimental results, which stimulated this paper. This work was supported in part by the National Science Foundation through grant No. DMR-81-4842 and through the Materials Research Laboratory at The Ohio State University.

*Address after 15 Sept. 1983: Department of Physics, University of California—San Diego, La Jolla, CA 92093.

[1]For many review articles and references up to 1979, see *Inhomogeneous Superconductors 1979, (Berkeley Springs, W.V.)*, Proceedings of the Conference on Inhomogeneous Superconductors edited by D. U. Gubser, T. L. Francavilla, J. R. Leibowitz, and S. A. Wolf (AIP, New York, 1979).

[2]M. Tinkham, in *Conference on Electrical Transport and Optical Properties of Inhomogeneous Media (Ohio State University, 1977)*, edited by J. C. Garland and D. B. Tanner (AIP, New York, 1978).

[3]G. Deutscher, O. Entin-Wohlman, S. Fishman, and Y. Shapira, Phys. Rev. B 21, 5041 (1980).

[4]S. A. Wolf, D. U. Gubser, and Y. Imry, Phys. Rev. Lett. 42, 324 (1979).

[5]S. A. Wolf, D. U. Gubser, W. W. Fuller, J. C. Garland, and R. S. Newrock, Phys. Rev. Lett. 47, 1071 (1982).

[6]D. J. Resnick, J. C. Garland, J. T. Boyd, S. Shoemaker, and R. S. Newrock, Phys. Rev. Lett. 47, 1542 (1981).

[7]C. J. Lobb, M. Tinkham, T. M. Klapwijk, and A. D. Smith, in *Low Temperature Physics—LT16*, edited by W. G. Clarke (North-Holland, Amsterdam, 1981), p. 17.

[8]R. Laibowitz, R. Broers, D. Stroud, and B. R. Patton, in Ref. 1, pp. 278—281.

[9]N. A. H. K. Rao, E. D. Dahlberg, A. M. Goldman, L. E. Toth, and C. Umbach, Phys. Rev. Lett. 44, 98 (1980).

[10]T. A. Worthington, P. Lindenfeld, and G. Deutscher, Phys. Rev. B 21, 5031 (1980).

[11]G. Deutscher, I. Grave, and S. Alexander, Phys. Rev. Lett. 48, 1497 (1982).

[12]For reviews of percolation theory, see, e.g., D. Stauffer, Phys. Rep. 54, 3 (1979); S. Kirkpatrick, Rev. Mod. Phys. 45, 374 (1973).

[13]J. P. Straley, J. Phys. C 9, 783 (1976); Phys. Rev. B 15, 5733 (1977).

[14]A. L. Efros and B. I. Shklovskii, Phys. Status Solidi B 76, 475 (1976).

[15]S. Kirkpatrick, in Ref. 1, p. 79.

[16]G. Deutscher, Y. Imry, and L. Gunther, Phys. Rev. B 10, 4598 (1974).

[17]B. R. Patton, W. Lamb, and D. Stroud, in Ref. 1, p. 13.

[18]C. Ebner and D. Stroud, Phys. Rev. B 23, 6164 (1981); 24, 5719 (1982).

[19]M. R. Beasley, J. E. Mooij, and T. P. Orlando, Phys. Rev. Lett. 42, 1165 (1979).

[20]B. I. Halperin and D. R. Nelson, J. Low Temp. Phys. 36, 599 (1979).

[21]S. Doniach and B. A. Huberman, Phys. Rev. Lett. 42, 1169 (1979).

[22]J. M. Kosterlitz and D. J. Thouless, J. Phys. C 6, 1181 (1973);

V. L. Berezinskii, Zh. Eksp. Teor. Fiz. 61, 1144 (1971) [Sov. Phys. JETP 32, 493 (1971)].
[23]See Ref. 12 above.
[24]D. A. G. Bruggeman, Ann. Phys. (Leipzig) 24, 636 (1935).
[25]R. Landauer, J. Appl. Phys. 23, 779 (1952).
[26]D. R. Karecki, G. L. Carr, S. Perkowitz, D. U. Gubser, and S. A. Wolf, Phys. Rev. B 27, 5460 (1983). For earlier work, see also G. L. Carr, Ph.D. dissertation, Ohio State University, 1982 (unpublished).
[27]D. C. Mattis and J. Bardeen, Phys. Rev. 111, 412 (1958).
[28]M. Tinkham, *Introduction to Superconductivity* (McGraw-Hill, New York, 1975), p. 69.
[29]We neglect proximity-effect coupling between the grains. Although such couplings undoubtedly exist in real samples, we believe that the present model should adequately describe many composites at very low temperatures, i.e., well below any possible vortex-unbinding phase transitions. However, our results are easily extended to finite T where quasiparticle absorption becomes important.
[30]D. Stroud and D. J. Bergman, Phys. Rev. B 25, 2061 (1982).
[31]C. J. Lobb and D. J. Frank, J. Phys. C 12, L827 (1979).