ac magnetic susceptibility, Meissner effect, and bulk superconductivity

R. A. Hein
Department of Physics, The Catholic University of America, Washington, D.C. 20064
(Received 5 November 1984; revised manuscript received 4 November 1985)

ac magnetic-susceptibility data for several $Ba(Pb_{1-x}Bi_x)O_3$ samples indicate behavior quite different from that usually regarded as "typical" for alloys and compounds. These results focused attention on the criteria for bulk superconductivity in general and in particular on two recently proposed criteria for deducing bulk superconductivity from zero-field, ac magnetic-susceptibility data. These are (1) the appearance of an "excessive loss" as measured by a maximum or "peak" in plots of the loss component of the complex ac magnetic susceptibility as a function of temperature and (2) an observed dependence of the magnitude of the "ac signal" upon the ac measuring frequency. A series of measurements were performed on elements and alloys in both singly and multiply connected geometries which clearly reveal that the recently proposed criteria are not applicable in general. In addition, the ability of normal-state electrodynamics to account for the frequency dependence of the magnitude of the "ac signal" and the observation of an "excessive loss" peak in all samples under suitable experimental conditions suggest that the above criteria are not valid ones. It is concluded that zero-field, ac magnetic-susceptibility data are insufficient for discerning bulk from nonbulk superconductivity and that such data should not be used as a basis for Meissner-effect measurements or discussions.

I. INTRODUCTION

During the past decade, numerous reports have appeared dealing with the observance of superconductivity in multicomponent, multiphase systems as well as in metastable phases prepared by unconventional techniques. Implicit in many of these reports is the belief that the observed superconducting transition is a bulk property of the major phase present in the sample. Current controversy regarding bulk versus nonbulk superconductivity in some of the Chevrel phase materials has once again focused attention on the relevancy of magnetic-induction measurements as a criterion for the occurrence of bulk superconductivity. A recent experimental paper by McCallum *et al.*[1] suggests that one can use measurements of the zero-field ac magnetic susceptibility to obtain data which allow one to discriminate between bulk and nonbulk superconductivity. Khoder's[2] recent theoretical treatment also suggests that such data can be used to discriminate bulk from nonbulk superconductivity.

These recent developments are in conflict with the conclusion of Hein and Falge,[3] who state that zero-field ac magnetic-susceptibility data provide an insufficient data base from which to deduce the occurrence of bulk superconductivity. Khoder's conclusion with regard to the behavior of the loss component of the complex ac magnetic susceptibility conflicts with accepted interpretations of such data.[4,5]

Motivated by these concepts,[1,2] a survey of the recent literature was performed which revealed (i) a frequent misuse of the phrase "A Meissner effect was observed. . ." and (ii) the apparent acceptance of two mutually exclusive points of view with regard to the relationship between zero-field ac magnetic-susceptibility data and the occurrence of bulk superconductivity.

Based on a perceived need to clarify the status of ac magnetic-susceptibility data as criteria for bulk superconductivity, a series of experiments were performed which hopefully will convince the reader that such criteria are not of general validity. The successful application of normal-state electrodynamics to account for the observed effects strongly suggest that the proposed criteria may, in fact, be invalid ones.

II. BACKGROUND

Ever since the discovery of the Meissner-Ochsenfeld effect,[6] commonly known as the Meissner effect, magnetic-induction methods have been used in (a) the search for new superconductors and (b) studies of the magnetic properties of known superconductors. Shoenberg[7] pioneered the use of the ac mutual inductance bridge technique, a technique of ever increasing popularity due to its experimental simplicity and versatility with regard to sample size and shape. This latter feature is of paramount importance for current studies of complex material systems.

In studies of the superconducting state, such bridge techniques allow one to monitor the temperature and magnetic field dependence of the sample's ac magnetic susceptibility; a complex quantity usually denoted as $\chi(T)=\chi'(T)-i\chi''(T)$. If measurements are performed with no externally applied dc magnetic field H present, it is the initial magnetic susceptibility denoted as $\chi_0(T)=\chi_0'(T)-i\chi_0''(T)$ which is of interest. When an applied dc magnetic field is present and if H is parallel to h_{ac}, where h_{ac} is the ac field of the primary winding, the quantity of interest is the differential ac magnetic susceptibility denoted as $\chi_H(T)=\chi_H'(T)-i\chi_H''(T)$. The importance of this distinction has been emphasized.[3,7]

Reports of the discovery of a new superconductor based on an observed diamagnetic shift in $\chi_0(T)$ will usually address the question of "bulk" superconductivity in one of two ways. Historically, the first way was one in which the magnitude of the observed diamagnetic shift is compared to that observed for a known bulk superconductor of the same size and shape. This approach assumes χ_0(normal)-χ_0(superconducting)$=\chi_0(N\text{-}S)$ is proportional to the volume of the superconducting regions. Such comparisons are the basis for estimates of the fractional volume of the sample which is superconducting; some typical examples are: (i) 30% of the sample (Eu-Mo-S) was superconducting,[8(a)] (ii) 95% of the sample (Ba-K-Pb-Bi-O) went superconducting,[8(b)] (iii) a value of χ_0 equal to $(93\pm7)\%$ of $-1/4\pi$ was observed[9] in $(Sn)_x$, and (iv) superconductivity is indeed a bulk phenomenon[10] in *bis*-tetramethyltetraselenafulvalene hexafluorophosphate $[(TMTSF)_2PF_6]$. The second way used to establish bulk superconductivity from measurements of magnetic properties is to state that a Meissner effect was observed. Recent examples of this approach are reports of (i) a Meissner effect in a polymeric system[9] and in composite systems;[11,12] (ii) the observation of an ac Meissner effect in an organic system,[10] (iii) a static Meissner effect[13] in $CuCe_2Si_2$, and (iv) a pressure-induced dc Meissner effect in a Chevrel-phase compound.[14]

The literature contains many reports by proponents of the school of thought who argue that conclusions concerning the occurrence of a Meissner effect or superconducting volumes from either the magnitude of $\chi_0(N\text{-}S)$ or initial slopes of magnetization curves are misleading or unwarranted. Hudson's very early work[15] showed that only a few percent of a second phase present as a superconducting impurity can produce large diamagnetic signals in dc mutual inductance measurements. In analyses of ac mutual inductance techniques the effects of "screening" current have been emphasized,[1,5] yet one still reads of divergent points of view. For example, Ott *et al.*[16] state, "the strong diamagnetic signal $\chi_0(T)$ alone gives no definite evidence for bulk superconductivity in UBe_{13}," while Harrison *et al.*[17] believe on the basis of $\chi_0(T)$ measurements that superconductivity in the Sn-Eu-Mo-S system is a bulk effect; Meng *et al.*[18] state in effect that $\chi_0(T)$ data "can provide an upper limit on the volume fraction of a superconducting transition." Clearly, both schools of thought cannot be correct.

What is the significance of the adjective ac in "an ac Meissner effect?" The use of qualifying adjectives such as dc or static with the phrase Meissner effect is misleading and/or incorrect as it implied the existence of more than one kind of Meissner effect. There is but one Meissner effect and it denotes the ability of a sample to expel, upon entering the superconducting state, all of the magnetic flux present when the sample was in the normal state. Expulsion of all the magnetic flux—the Meissner effect, or in the parlance of some, the ideal Meissner effect, is a rare phenomenon only occurring with physically and chemically pure samples. Reports of a Meissner effect—a hallmark of bulk superconductivity—must be viewed with considerable skepticism in cases where flux expulsion measurements were not performed.

The phrase ac Meissner effect is a misnomer as it describes perfect magnetic shielding or flux exclusion and not expulsion of magnetic flux. A sample exhibiting a Meissner effect will also exhibit perfect shielding but not vice versa, for example, Ribault *et al.*[10] state that their $\chi_0(T)$ data indicate a complete ac Meissner effect yet the $\chi_H(T)$ data failed to exhibit a differential paramagnetic effect (DPE) hence little or no flux expulsion took place.[3] To associate the Meissner effect with $\chi_0(N\text{-}S)$ data obtained by ac induction techniques, places perfect ac shielding on an equal footing with flux expulsion, which it is not! The deduction of a dc Meissner effect from the initial slope of a virgin magnetization curve is also misleading and/or incorrect[14] as here too, one only observes the result of perfect shielding.

The availability of SQUID (superconducting quantum-interference device) magnetometers has resulted in an increase in flux expulsion experiments.[1,13,19,20] In general such measurements are performed on materials already reported to be superconductors and are not well suited for "survey" type experiments involving newly synthesized materials. Given that one is using an ac inductance bridge technique to detect the superconducting transition temperature T_c, one has a ready means to comment on flux expulsion. All one need do is complement the $\chi_0(T)$ data with $\chi_H(T)$ data. If one observes a field-induced peak[3] in $\chi_H(T)$, the DPE, then he knows the sample is expelling flux. Whether or not $\chi_H(T)$ data allow one to establish a lower bound on the superconducting volume, as is claimed[13] for SQUID magnetometry, is yet to be ascertained.

Acceptance of "bulk" superconductors only those alloys, compounds, etc. that exhibit nearly complete flux expulsion would result in very few bona fide bulk superconductors. Such a criterion is much too restrictive. A partial Meissner effect, giving evidence of some flux expulsion, is clear proof that "bulk" superconductivity has occurred in some non-negligible volume of the sample.

Khoder's[2] assertion that a peak in $\chi_0''(T)$ at $T\approx T_c$ is proof of bulk superconductivity, or the claim of McCallum *et al.*[1] that measurements of $\chi_0(N\text{-}S)$ as a function of frequency can discriminate bulk from nonbulk superconductivity, if substantiated, would be very helpful indeed in the establishment of a working criterion for deducing the occurrence of bulk superconductivity from $\chi_0(T)$ data. The present study was undertaken with three goals in mind: (i) to present quantitative data that one should not deduce bulk properties from $\chi_0(N\text{-}S)$, (ii) to investigate the frequency dependence of $\chi_0(N\text{-}S)$ as a means of discriminating between bulk and nonbulk superconductivity and (iii) to study the peak in $\chi_0''(T)$ in order to discern between various conflicting models which ascribe this peak to either filamentary superconductivity,[4] bulk superconductivity,[2] or normal state behavior.[5]

III. EXPERIMENTAL DETAILS

Temperatures as low as 1.3 K are attained by the use of conventional glass Dewars and liquid helium. A room temperature magnetic shield is used to reduce the ambient magnetic field to <15 mOe. When required, an external magnetic field up to 200 Oe is supplied by a liquidnitrogen-cooled copper solenoid wound directly on the helium Dewar. A room-temperature "loader" of the type described by Das *et al.*[21] permits one to cycle the sample holder and its addendum, i.e., germanium thermometer, heater, etc. between room temperature and helium temperature in about two hours or less with a minimal consumption of liquid helium. A Hartshorn-type bridge[22] incorporating a two-phase lock-in analyzer [Princeton Applied Research (PAR) Model 5204] is used to monitor temperature-induced changes in the mutual inductance $M = M' - iM''$ of a copper coil system consisting of a coaxial pair of oppositely wound secondary coils spaced 1.5 in. apart and a common concentric primary winding. Each secondary is 0.625 in. long and contains 7,500 turns of AWG (American wire gauge) No. 38 copper wire. The primary is 5 in. long and contains 1,845 turns of AWG No. 38 copper wire.

The brass coil form of the secondary coils is an integral part of the vacuum system (0.625-in. i.d.) of the room-temperature loader. These coils remain at liquid-helium temperatures throughout a given experiment. With a sample located at the center of one of the secondary windings, any change in the mutual inductance of the coil system accompanying a change in temperature of the sample and holder is related to the change in the ac magnetic susceptibility of the sample, provided the susceptibility of the sample holder is essentially constant over the narrow temperature interval of interest.

Balancing of the mutual inductance bridge is initially accomplished with the sample in either the superconducting or normal state. A substantial imbalance in the reactive component M' is then introduced by means of a ratio-tran; any accompanying imbalance in the resistive component M'' is brought back to zero by adjusting the phase of the lock-in analyzer. Returning the ratio-tran to the balance setting should produce no imbalance in M''. If this condition is met, the quadrature channel of the lock-in responds only to changes in M', the reactive component of M, and the in-phase channel responds only to changes in M'', the loss or resistive component.

$M(T)$ data are obtained in two ways:[3] (i) point-by-point method—here the temperature is held constant at fixed intervals and the bridge is rebalanced at each temperature setting. Any changes in M' and M'' which accompany changes in the temperature are proportional to changes in $\chi'(T)$ and $\chi''(T)$ of the sample. (ii) Temperature sweep method—with the bridge initially in the balanced condition, any imbalance in the output voltage of the bridge (in- and out-of-phase voltages) resulting from a monotonic change in temperature is detected and amplified by the lock-in analyzer. The rectified outputs of the amplifier are displayed on the Y axis of a two-pen recorder. The dc voltage developed across a germanium resistor thermometer is detected by a differential voltmeter the output of which drives the X axis of the recorder. In this manner continuous tracings of the in- and out-of-phase voltages V_R and V_Q as functions of temperature are obtained. For given values of h_{ac} and ac measuring frequency $V_Q(T) \approx M'(T) \approx \chi'(T)$ and $V_R(T) \approx M''(T) \approx \chi''(T)$. The temperature sweep method, being the less tedious of the two methods, is the one most commonly employed in the search for new superconductors.

The majority of samples used in this study were cylinders of the same nominal dimensions; 4.5 mm in diameter and 4.8 mm in length. Data obtained on solid cylinders of Pb, Sn, and Pb-Bi were complemented with data obtained for two composite cylinders; these are brass cylinders one of which has a 0.05-mm coating of solder on its sides (ends are not plated) while the other has a tin filled groove (0.8 mm wide and 0.8mm deep) around its center section. Samples of Nb, TiCr, V_3Ga, and $BaPb_{1-x}Bi_xO_3$ were also investigated.

IV. RESULTS AND DISCUSSION

A. Magnitude of $\chi'_0(N\text{-}S)$

Zero-field transitions for five samples as determined by the temperature-sweep method are depicted in Fig. 1.[23] The data consist of recorder tracings of $V_Q(T)$, the out-of-phase voltage, as a function of the reduced temperature. The bridge was balanced with the samples in the normal state and $V_Q(N\text{-}S) = V_Q(N) - V_Q(S) \approx \chi'_0(N\text{-}S)$ is the unbalanced voltage resulting from the change in the sample's magnetic susceptibility as it enters the superconducting state. To facilitate comparison, the abscissa is a reduced temperature $t = T/T_c$, where T_c is defined as the midpoint of the $V_Q(N\text{-}S)$ trace.

Balancing the bridge with the sample in the normal state results in $V_Q(N) = 0$. However with the amplifier gain set at 10^5, it was noted that samples D and E produced small overloads of the output meter (± 1 V). For these cases, it was found expedient to use the ratio-tran to produce a small negative imbalance so that $V_Q(N) \neq 0$; in this way one could keep a gain of 10^5 and record output voltages as large as 2 V. Thus the ordinate is labeled $\Delta V_Q(N\text{-}S)$ with $V_Q(N) = 0$ for samples A, B, and C.

The data of Fig. 1 show that samples A and B (solid Sn cylinder and Sn ring) yield almost identical $V_Q(N\text{-}S)$ values whereas the volumes of Sn present in the two sam-

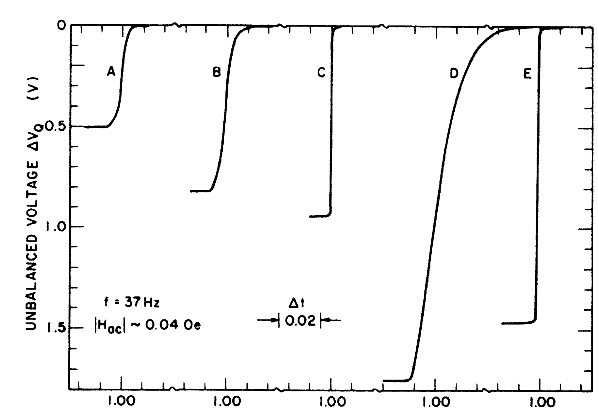

FIG. 1. Recorder traces of the output voltage of the quadrature channel of the lock-in amplifier (gain $= 10^5$) as a function of the appropriate reduced temperature for (A) Sn cylinder, (B) Sn ring, (C) Pb cylinder, (D) "Pb" hollow cylinder, and (E) Pb-Bi(10 at. %) cylinder. The ΔV_Q notation is explained in Sec. IV A of the text.

ples differ by an order of magnitude. The values of $V_Q(N\text{-}S)$ for samples C and D (Pb cylinder and solder-plated brass cylinder) are comparable yet the volumes of superconducting material differ by two orders of magnitude.

These data demonstrate in a quantitative manner, the basic unsoundness of estimating fractional volumes of superconducting material present in a sample from relative values of $V_Q(N\text{-}S) \approx \mathcal{X}_0(N\text{-}S)$. Based on elementary expressions for the mutual inductance of coils and induced voltages, one finds for the sample coil configuration used in this study that

$$V_Q(N\text{-}S) = \Delta M'(N\text{-}S)\,dI/dt = I\omega\,\Delta M'(N\text{-}S)\ ,$$
$$\Delta M'(N\text{-}S) \approx a(\delta - \lambda)\ , \tag{1}$$

where a is radius of the cylindrical sample, δ is the normal state skin depth, λ is the superconducting penetration depth, and ω is 2π times the frequency f of the ac measuring field h_{ac} produced by the primary current I.

This expression relates $V_Q(N\text{-}S)$ to the difference in the cross sections for shielding of the ac magnetic field when the sample is normal and when it is superconducting. Clearly $\Delta M'(N\text{-}S)$, hence $V_Q(N\text{-}S)$, reflects these effective cross sections, not the volume of superconducting material present in the sample. An awareness of this has led to the use of powdered samples.[1,24] Powder samples are less affected by eddy currents and decrease the effects of multiply connected superconducting regions.

The above analysis shows that if λ does not vary significantly compared to the δ values, then samples with the larger δ will yield the larger $\Delta M'(N\text{-}S)$. The data of Fig. 1 indicate $\delta(\text{Pb}) > \delta(\text{Sn})$. Since $\delta^2 = 10^9\rho/4\pi^2 f$, where δ is in units of cm if ρ is in units of Ω cm and f is in sec^{-1}. These data suggest that $\delta(\text{Pb}) > 3\delta(\text{Sn})$. It is the role of δ in determining $\Delta M'(N\text{-}S)$, noted by Shoenberg[7] and discussed in reports dealing with the magnetic response of known superconductors to applied ac magnetic fields,[5,25] which is overlooked when one uses the magnitude of $V_Q(N\text{-}S)$ to estimate superconductivity volumes. The above relationship between $\Delta M'(N\text{-}S)$ and δ leads quite naturally to a consideration of $V_Q(N\text{-}S)$ as a function of the measuring frequency.

B. Frequency dependence of $V_Q(N\text{-}S)$

In their study of the EuMo_6S_8 system, McCallum *et al.*[1] emphasized that ac magnetic-susceptibility data yield information about the effects of screening currents induced on the surface of the sample and not about the Meissner effect. They also state that (i) for a bulk superconductor the screening of the sample is complete and independent of the frequency and (ii) if the observed signal is due to superconducting loops on the surface or along grain boundaries "the screening is less effective at low frequencies and the signal decreases as the frequency is reduced." Thus by plotting the ratio of the "ac signals" of two samples, one of which is known to be a bulk superconductor, one can in principle discern bulk from nonbulk superconductivity.

Figure 2 contains plots of $V_Q(T)$ for solid cylinders of Pb and of a $\text{Pb}_{0.9}\text{Bi}_{0.1}$ alloy as functions of temperature

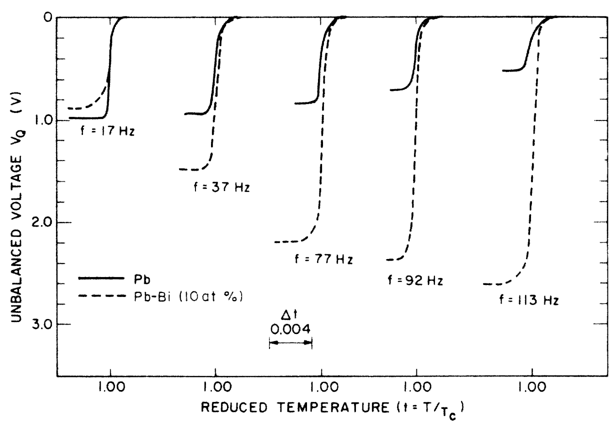

FIG. 2. Recorder traces of the outout voltage of the quadrature channel of the lock-in amplifier (gain $=10^5$) as a function of the reduced temperature for various values of the ac measuring frequency; $h_{ac} = 0.04$ Oe (rms). The tiaces for Pb were as shown, for Pb-Bi(10 at. %) the 37 to 92 Hz data are scaled from a gain of 4×10^4.

for several values of the measuring frequency. These data, obtained with an ac measuring field of 0.04 Oe (rms) and with the bridge balanced when the samples were in the normal state, display two obvious features: (i) $V_Q(N\text{-}S)$ decreases with increasing frequencies for Pb while it increases with increasing frequencies for the Pb-Bi alloy and (ii) the ratio of $V_Q(N\text{-}S)^{\text{Pb-Bi}}/[V_Q(N\text{-}S)]^{\text{Pb}}$ increases from about 0.9 at 17 Hz to about 5 at 113 Hz; values in close agreement with those reported by McCallum *et al.*[1]

Since both Pb and $\text{Pb}_{0.9}\text{Bi}_{0.1}$ are believed to be bulk superconductors, a check of the ac bridge was made in order to rule out any experimental peculiarity as the cause of the frequency-dependent ratio. Incorporating a variable inductometer in the ac bridge circuitry showed that over the frequency range of interest the relationship between V_Q and $\Delta M'$ is linear in f in agreement with Eq. (1). Thus the frequency dependence of the above ratio must reside in different frequency dependences of $\Delta M'(N\text{-}S$ for Pb and the Pb-Bi alloy. In the case of perfect diamagnetism one has the following expression:

$$\mathcal{X}_0'(S) = \frac{1}{1-n}\left[\frac{dm(H,T)}{dh}\right]_{H=0} = \frac{1}{1-n}\left[\frac{-1}{4\pi}\right]\ , \tag{2}$$

where n is the sample's demagnetization factor, $m(H,T)$ is the magnetic moment of the sample, and H is the longitudinal dc magnitude field. Ignoring any time effects, one sees that $\mathcal{X}_0'(S)$ is independent of the frequency; consequently so is $M'(S)$. Therefore any frequency dependence in $\Delta M'(N\text{-}S) = M'(N) - M'(S)$ must reside in $M'(N)$.

The above discussion is just a restatement of the remark of McCallum *et al.*[1] that the shielding of a bulk superconductor is complete and independent of the frequency; however, the "ac signal" is the difference in the ac shielding between that of the superconducting state and the normal state. This difference will only be frequency independent provided the shielding in the normal state [or $\chi_0'(N)$] is independent of the frequency.

Based on Eq. (1), one sees that $M'(N) \approx \delta_n \approx f^{-1/2}$, hence $V_Q(N\text{-}S) \approx f^{1/2}$. The Pb-Bi alloy data is in agreement with such a frequency dependence. The Pb data do not follow this simple relationship. In fact $V_Q(N\text{-}S)$ for Pb is nearly constant for frequencies between 17 and 37 Hz and then decreases at the higher frequencies. This behavior requires a $\Delta M'(N\text{-}S)$ which varies like f^{-1} at the low frequencies and more strongly at the higher frequencies.

The nearly constant $V_Q(N\text{-}S)$ value for 17 Hz $< f <$ 37 Hz in Fig. 2 prompted a more detailed study in the low-frequency regime (i.e., $f <$ 70 Hz). In order to rule out the possibility that this observed dependence was peculiar to the sample and not a property of Pb *per se* a second Pb sample was machined to the "exact" dimensions of the Pb-Bi cylinder. Once again a frequency-independent $V_Q(N\text{-}S)$ was observed at the lower frequencies. Figure 3 displays the frequency dependence of $V_Q(N\text{-}S)$ in terms of the measured $\Delta M'(N\text{-}S)$. Included in Fig. 3 are data for the first Pb cylinder, the Pb-Bi cylinder, a Nb cylinder, and a "cylindrical" sample of nominal V_3Ga. Note that if one compares the $M'(N\text{-}S)$ values obtained for Pb-Bi to those of Pb one obtains a frequency-dependent ratio as noted by McCallum *et al.*;[1] replacing the Pb values with values obtained for the Nb cylinder yields a ratio that is nearly independent of the frequency. Thus it appears that if one compares the "ac signal" of a superconductor to that of another superconductor of higher normal-state electrical conductivity one will observe a frequency-dependent ratio of the kind reported by McCallum *et al.*[1] However, if the "bulk" superconductor has a normal-state electrical conductivity not much larger than that of the alloy the ratio will be nearly frequency independent.

These observations lead one to believe that the frequency-dependent ratio of the ac signal is a consequence of differences in the normal-state properties (see Sec. IV C) and not a consequence bulk versus nonbulk superconductivity *per se*. Thus one cannot use such an observation as a criterion for bulk versus nonbulk superconductivity; one needs additional information such as the effect of "powdering" upon the magnitude of the diamagnetic signal.[1,24]

C. Peaks in $V_R(T) \approx \chi_0'(T)$

Another aspect of ac magnetic-susceptibility data that has been used to discriminate between bulk and filamentary superconductors is the behavior of $\chi_0''(T) \approx M''(T)$. It has been suggested that the peak in $\chi_0''(T)$ which accompanies the N-S transition in alloys and compounds both is[2] and is not[4,5,23] a bulk phenomenon. The postulate that the peak in $\chi_0''(T)$ at $T \gtrsim T_c$ is due to the presence of isolated superconducting filaments with T_c values in excess of the bulk matrix was the subject of considerable controversy.[26] Data of Fig. 4, obtained at a frequency of $f = 37$ Hz, show that Sn and Pb do not exhibit a peak in $V_R(T) \approx \chi_0''(T)$ but that Pb-Bi(10 at. %) as well as samples B (Sn ring) and D (hollow cylinder) do show peaks with the peak in the Pb-Bi data being the sharpest. Based on the criterion of Maxwell and Strongin,[4] one would conclude that Sn and Pb are bulk superconductors while Pb-Bi, sample B and sample D are filamentary superconductors. Khoder's criterion[2] leads one to the opposite conclusions. One may state with little fear of criticism that neither criterion is universally applicable.

The observation of peaks in $V_R(T)$ data for the multiply connected geometries (Fig. 4) is believed to be the result of "alloying" which occurred in their fabrication and not a result of their geometrical configurations. This is based on the fact that a machined hollow cylinder of Pb (0.5 mm wall thickness) yielded results quite similar to that of the solid Pb cylinder, i.e., no peak in $V_R(T)$ for frequencies > 37 Hz.

Maxwell and Strongin based their explanation of the appearance of peaks in $\chi_0''(T)$ for strained or impure sam-

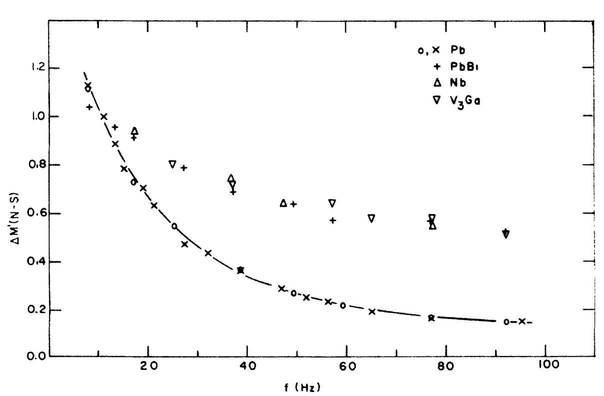

FIG. 3. Differences in M' values obtained by balancing the bridge with the samples of Fig. 1 in the normal and superconducting states, i.e., $\Delta M'(N\text{-}S)$ as a function of the ac measuring frequency. Data are normalized to unity at $f = 11$ Hz.

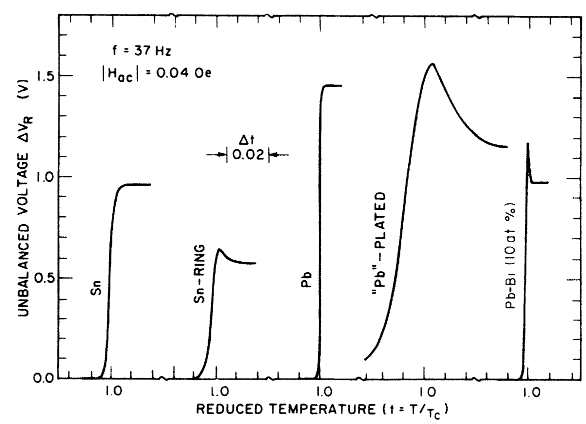

FIG. 4. Recorder traces of the output voltages of the in-phase channel of the lock-in amplifier (gain $= 10^5$) as a function of the appropriate reduced temperature for the five samples of Fig. 1. The ΔV_R notation is explained in Sec. IV A of the text.

ples as well as the absence of such peaks in pure, unstrained samples on the normal-state electrodynamics of Landau and Lifshitz.[27] These authors[27] treated the case on an infinitely-long, normally conducting cylinder of radius a, in a longitudinally applied ac magnetic field and presented asymptotic formulas for χ' and χ'' as a function of a/δ; Maxwell and Strongin used the results of Landau and Lifshitz to obtain the behavior shown in Fig. 5.

Based on the χ'' dependence shown in Fig. 5, the observation of a peak in $\chi_0''(T)$ at the N-S transition requires[4] that two conditions be met: (i) the normal-state skin depth at $T>T_c$ must have a value such that $a/\delta<1.8$, and (ii) as one approaches the N-S transition from $T>T_c'$, δ must decrease so as to shift a/δ to values in excess of 1.8. It was in order to have the required shift in a/δ take place before the full diamagnetic shielding of the superconducting state occurs, that Maxwell and Strongin proposed their filamentary model. In this model, as the filaments become superconducting, the effective conductivity of the sample is increased by an amount sufficient to cause a/δ to increase to values in excess of 1.8. This shift will occur at temperatures slightly higher than the T_c of the bulk material as the T_c of the filaments is postulated to be slightly higher than that of the bulk. Belief in this model has led to the use of a peak in $\chi_0''(T)$ or $V_R(T)$ as a hallmark of filamentary superconductors.[28]

Khoder's mathematical treatment,[2] based on current microscopic theories of the superconducting state, identifies the cause of this extra loss, i.e., χ_0'' (peak) $>\chi_0''$ (normal), as the extra kinetic energy required to accelerate the supercurrent, and hence is a property of bulk superconductors. An opposite point of view is expressed by Gregory[5] who treats the eddy current model in considerable detail and concludes that a peak in $\chi_0''(T)$ is a normal-state phenomenon and does not reflect properties of the superconducting state *per se*. He shows that a peak in $\chi_0''(T)$ will occur provided the measuring frequency is less than some critical frequency f_c, where

$$f_c=(1.8/a)^2(10^9\rho/4\pi^2)\ . \tag{3}$$

Assuming a value[7] for ρ of 3×10^{-9} Ω cm for machined Pb and $a=0.22$ cm, the above formula leads to a value of $f_c=5$ Hz. Figure 6, shows $V_R(T)\approx\chi_0''(T)$ data obtained at several values of the frequency for the Pb and $Pb_{0.9}Bi_{0.1}$ cylinders. A peak in $V_R(T)$ for Pb is clearly evident provided $f<17$ Hz; an f_c of 17 Hz requires $\rho=10\times10^{-9}$ Ω cm a reasonable value for machined Pb. The data of Fig. 6 tend to support the idea of Gregory and his eddy current model for the behavior of type-I superconductors.

The eddy current model also provides a qualitative explanation for the frequency dependence of $V_Q(N$-$S)$ and $\Delta M'(N$-$S)$ shown in Figs. 2 and 3. According to the behavior depicted in Fig. 5, $\chi'(N)$ is a monotonic function of a/δ and is an appreciable fraction of $\chi'(S)$ for $a/\delta>1$. In order to have $\chi_0'(N)$ be negligible with respect to $\chi_0'(S)$ one must use a measuring frequency corresponding to an a/δ value of 0.5 or less. In this case the magnitude of $\Delta M'(N$-$S)$ will be essentially independent of the measur-

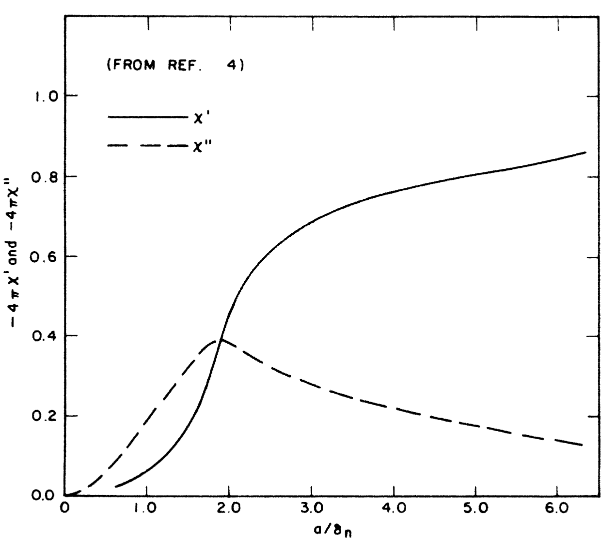

FIG. 5. In-phase (χ') and out-of-phase (χ'') components of the complex ac magnetic susceptibility of a normally conducting cylinder of infinite length and radius a as a function of a/δ_n; δ_n is the normal-state skin depth.

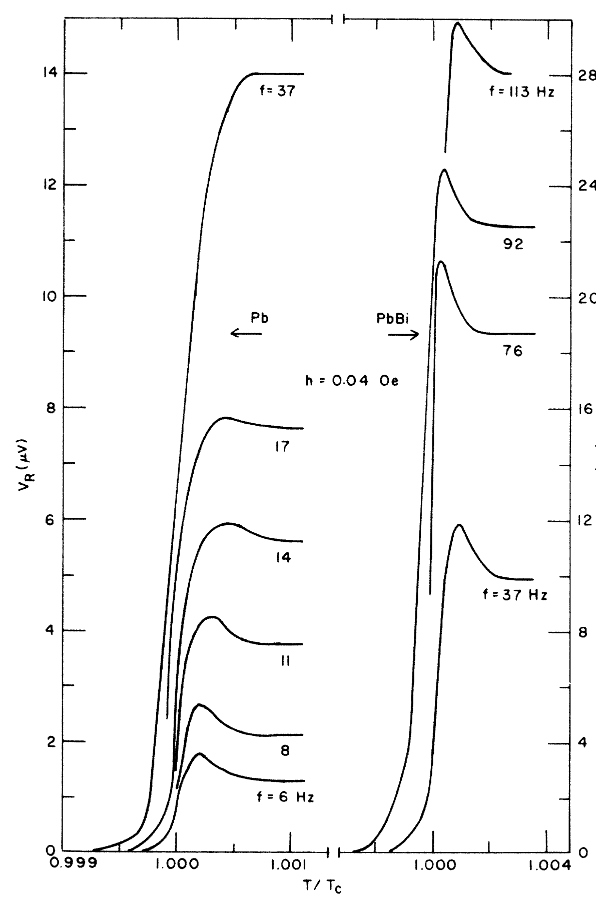

FIG. 6. V_R and Pb and Pb-Bi(10 at.%) as functions of reduced temperature for several values of the ac measuring frequency for an ac field strength of 0.04 Oe (rms). The ordinant is expressed in terms of the input voltage to the in-phase channel of the lock-in amplifier.

ing frequency and have its maximum value, i.e., $\chi'_0(S)$. Since a is less than δ, Eq. (1) becomes $\Delta M'(N\text{-}S) = a(a-\lambda)$, which is also independent of the measuring frequency.

Data shown in Fig. 3 show that a frequency-independent $\Delta M'(N\text{-}S)$ is not observed. For the Pb samples used in this study, $\Delta M'(N\text{-}S)$ was still increasing with decreasing frequency down to 8 Hz, the lowest frequency used in this study. The same situation applies to $Pb_{0.9}Bi_{0.1}$, V_3Ga, and Nb. For the latter sample, 11 Hz was the lowest frequency used.

From the study of $V_R(T) \approx \chi''(T)$ as a function of frequency, a value of 17 Hz was assigned to f_c for Pb, from which a value of $\rho = 10\times 10^{-9}\ \Omega\,\text{cm}$ was deduced. Incorporating these values of ρ and f_c into the eddy current model shows that the frequency range of 8–60 Hz corresponds to a/δ values ranging from 1.2–3.4 which, according to the data of Fig. 5, is just the range of a/δ over which $\chi'(N)$ is strongly increasing with increasing a/δ or frequency. Therefore one can expect $\Delta M'(N\text{-}S) \approx \chi'_0(N) - \chi'_0(S)$ to decrease sharply with increasing frequency at these low frequencies. In order to account for the factor of 3 decrease in $\Delta M'(N\text{-}S)$ in Fig. 3, a slight modification of the $\chi'(N)$ curve in Fig. 5 is required. The data as shown will lead to a decrease in $\Delta M'(N\text{-}S)$ between 8 and 60 Hz of about a factor of 4. The fact that the cylinders used in this work have nonzero demagnetization factors ($n \approx 0.2$) may account for this discrepancy.

The variation of $\chi'(N)$ and $\chi''(N)$ with a/δ which results from the application of normal-state electrodynamics (Fig. 5) can account for the observed frequency dependence of $\Delta M'(N\text{-}S)$ and the existence of a critical frequency for a loss peak in $V_R(T) \approx \chi''_0(T)$. Thus one may argue that the loss peak and f_c observed in the N-S transition is[5] a property of the normal state (see Sec. IV D).

Curves a and a' of Fig. 7 are typical of the results obtained for $BaPb_{0.76}Bi_{0.24}O_3$. These data were obtained with a "composite" sample consisting of four irregularly shaped pieces of single crystals ($d \approx 1$ mm). Consequently, the rather broad N-S transition is not surprising. However, the fact that $V_R(T)$ is monotonic at all frequencies (11–113 Hz) is a disturbing result. Given that $BaPb_{0.76}Bi_{0.24}O_3$ is a high resistivity material ($\rho > 10^{-4}\ \Omega\,\text{cm}$) the experimental conditions yield $a/\delta < 1.8$. Hence one would expect[4,5] a peak in the $V_R(T)$ data. According to Hein and Falge,[3] the absence of a peak in $V_R(T)$ implies little or no flux trapping, a feature inconsistent with the rather broad transition and proven not to be the case when $V_Q(H,T)$ data showed no evidence of a DPE.[3] Khoder's criterion[2] classifies this compound as a nonbulk superconductor.

$V_R(T)$ data obtained for a cylindrical sample of $TiCr_{1.65}$ showed a peak in agreement with published results.[23] It is noted in passing that the earlier work on $TiCr_{1.65}$ used the point-by-point method, Sec. III, and the peak was more pronounced. Such data reaffirm the belief that broad transitions, *per se*, do not prevent the observation of a peak in $V_R(T)$. Results obtained with a nominal V_3Ga sample, known[21] to have a concentration gradient in the surface layer showed three distinct peaks in $V_R(T)$; thus inhomogeneities *per se* cannot account for the absence of a peak in the a curve of Fig. 7.

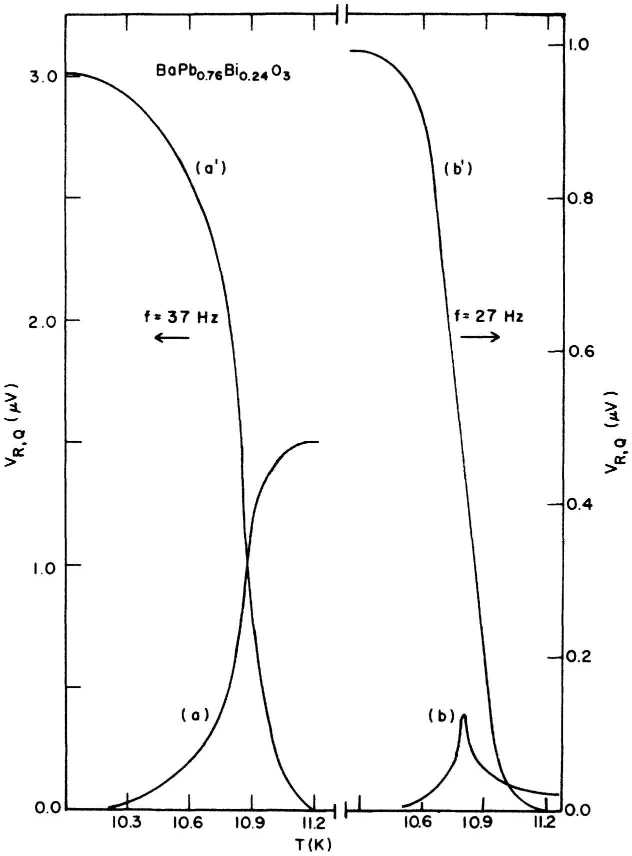

FIG. 7. V_R (curves a and b) and V_Q (curves a' and b') as functions of temperature for two "samples" (see Sec. IV C of the text) of $BaPb_{0.76}Bi_{0.24}O_3$. The ordinant is expressed in terms of the input voltages to the lock-in amplifier. For clarity, V_Q has been inverted.

To investigate possible demagnetization effects associated with the composite nature of the $BaPb_{0.76}Bi_{0.24}O_3$ sample, a mutual induction coil of smaller inside diameter (0.375 in.) was used to measure the ac magnetic susceptibility of just one of the four single-crystal pieces of $BaPb_{0.76}Bi_{0.24}O_3$ (volume $\approx 3.4\ \text{mm}^3$). Whereas no discernible peak in $V_R(T)$ was evident with $h_{ac} = 0.07$ Oe, increasing the magnitude of h_{ac} to 0.25 Oe results in data shown as curves b and b' in Fig. 7. With this larger value for h_{ac}, peaks were evident at all frequencies (11–92 Hz), a result which suggests that one needs to exceed some minimal value of h_{ac} in order to produce a peak in $V_R(T) \approx \chi''_0(T)$. Eddy current model considerations do not involve a minimum h_{ac}; the only prerequisite is that one has the required sensitivity to detect changes in $\chi''_0(T)$. This requirement is clearly fulfilled if one detects a reasonable $\chi''_0(N\text{-}S)$ signal.

A study of $V_R(T)$ as a function of h_{ac} was not performed for $BaPb_{0.76}Bi_{0.24}O_3$ compound, as the primary objective of this part of the investigation concerned the occurrence of the peak and the criterion of Khoder.[2]

The above results, which suggest that the observation of a peak in $V_R(T)$ is dependent on the appropriate choice of h_{ac} and f, led to a detailed study of the effect of the magnitude of h_{ac} upon the $V_R(T)$ response of the Nb cylinder used to obtain the $\Delta M'(N\text{-}S)$ data of Fig. 3. Whereas the $V_Q(T)$ data for this cylinder behaved in a manner consistent with that of the other elements and alloys, it displayed an abnormal $V_R(T)$ dependence; that is, with $h_{ac} \approx 0.04$ Oe no peaks were observed at 11 or 37 Hz while small peaks were observed at $f=77$ and 92 Hz. This result conflicts with the f_c prediction of the eddy current model[5] and stands in sharp contrast to the frequency dependence found in Pb, Fig. 6.

Figure 8 shows $V_R(T)$ for Nb as a function of h_{ac} at a fixed frequency of 19 Hz. Clearly the peak height, measured from the normal-state value, is a function of h_{ac} and the extra-loss peak exhibits structure. For the smaller values of h_{ac}, the structure is of a double-peak nature, and as h_{ac} increases in magnitude the lower temperature peak increases relative to the higher temperature peak. At the higher values of h_{ac} one observes a single-peak structure with a high-temperature "shoulder."

Reexamination of the earlier ($h_{ac} \approx 0.04$ Oe) data indicated that the 11-Hz data did show some small structure in $V_R(T)$ which had been dismissed as noise; however, the 37 Hz data clearly failed to exhibit a peak in $V_R(T)$. This fact prompted a study of the "peak" as a function of frequency. A value of h_{ac} equal to 0.36 Oe was chosen for which the data of Fig. 8 exhibits a relatively large single-loss peak with a high-temperature shoulder. Data of Fig. 9 reveal an unexpected result in that one sees, with increasing frequency, the relative decrease of the low-temperature peak, the reappearance of the double-peak structure, and the subsequent low-temperature shoulder. The relative decrease with increasing frequency in the overall peak height, with regard to the normal-state loss, is reminiscent of the f_c prediction of Gregory[5] (see remarks in Sec. IV D).

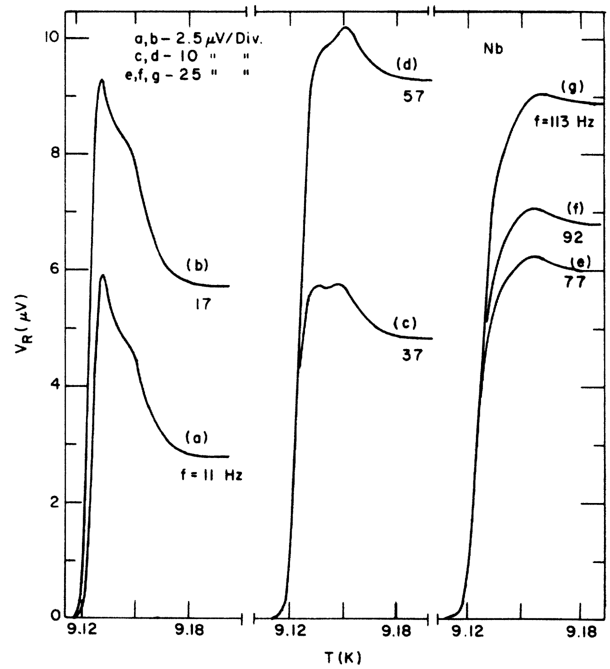

FIG. 9. V_R for the Nb cylinder as a function of temperature for several values of the ac measuring frequency for an ac field strength of 0.31 Oe (rms). The ordinant is expressed in terms of the input voltage to the in-phase channel of the lock-in amplifier; note changes in the scale for the ordinant.

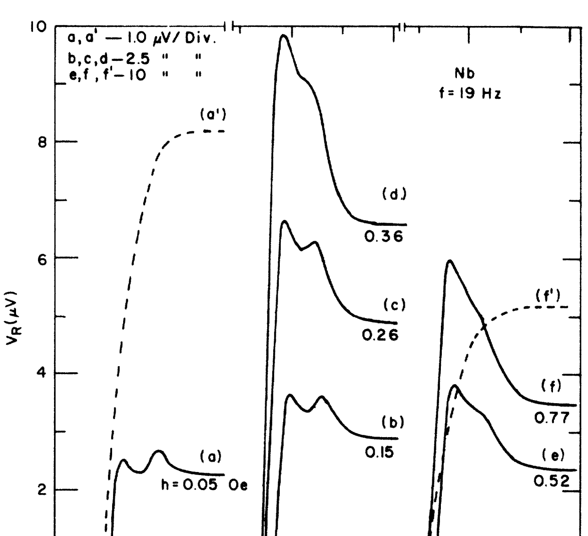

FIG. 8. V_R (solid curves) for the Nb cylinder as a function of temperature for several values of the ac magnetic field between 0.05 and 0.77 Oe (rms). V_Q (dashed curves) is shown for two values of h_{ac}. The ordinant is given in terms of input voltages to the lock-in amplifier; note changes in the scale for the ordinant.

On the basis of the $V_R(T)$ data presented in this section, it is concluded that a peak in $V_R(T)$ is ubiquitous to all superconducting transitions provided one uses an appropriate set of values for h_{ac} and f. Thus it is felt that the criterion proposed by Khoder[2] to discriminate between bulk and nonbulk superconductivity is not valid.

D. Peaks in $\chi'_H(T)$ and $\chi''_H(T)$: Loss mechanisms

Although this subject lies outside the stated goals of this work, a few remarks are in order. For the Pb solid cylinder, peaks are observed in both $\chi'_H(T)$ and $\chi''_H(T)$ (similar results for the Sn cylinder). The peak in $\chi'_H(T)$, referred to as the DPE is a "bulk" intermediate state effect[3,7] and so is the peak in $\chi''_H(T)$ when it accompanies a peak in $\chi'_H(T)$. When this is not the case, a peak in $\chi''_H(T)$ occurs in the high magnetic field portion (tail) of a highly irreversible magnetization curve. This has led[29] to the identification of the peak in $\chi''_H(T)$ with the value of $H_{c3}(T)$, the surface sheath critical magnetic field—a widely used criterion. While the utility of using the peak in $\chi''(H,T)$ to determine $H_{c3}(T)$ has been accepted, the mechanism which gives rise to this peak has been the subject of considerable discussion. Magnetic hysteresis models[30–32] have met with considerable success and Doidge *et al.*[33] showed that the peak in $\chi_0''(T)$ (Sec. IV C) is just the $H=0$ limit of the $\chi_H''(T)$ peak—thus one can argue that both peaks are due to flux trapping.

Cody and Miller[34] pointed out some shortcomings of the magnetic hysteresis model and developed an eddy current model. Their work preceded that of Gregory[5] and was based on the electrodynamics of Landau and Lifshitz.[27] Since both works[5,34] are based on the same concept, one again notes that peaks in $\chi_0''(T)$ and $\chi_H''(T)$ arise from the same cause. Gregory's remarks were confined to ideal type-I superconductors and stressed that peaks in $\chi_0''(T)$ should appear at sufficiently low frequencies (Sec. IV C).

Cody and Miller worked with thin Pb films (100 Å to 16 000 Å) and had to modify the expression of Landau and Lifshitz since the thin films were disk-shaped samples and not infinitely long cylinders. Their expression for the critical frequency replaces a with $\sqrt{ad}$, where a is the radius of the disk and d is the film thickness. In all aspects this treatment parallels that of Gregory for $\chi_0''(T)$.

Cody and Miller also conclude that a peak in $V_R(H,T)\approx\chi_H''(T)$ will be observed provided $f<f_c$. They argued that if one observes a peak in $V_R(H,T)$, one may calculate a value for an effective resistance ρ^*, which gives rise to this peak, i.e.,

$$\rho^*=\rho(\text{peak})=(\sqrt{ad}/1.8)^2(4\pi^2 f/10^9)\ .$$

Their observation that a 500-Å film of Pb with a radius of 0.5 cm showed a peak in $V_R(H,T)$ with $f=100$ Hz led them to estimate that $\rho^*\approx 3\times10^{-12}\ \Omega\,\text{cm}$ ($\ll\rho_n$). Cody and Miller speculated that flux-flow resistance could be used as a loss mechanism leading to peaks in $V_R(H,T)$.

Data shown in Fig. 10 pose a dilemma for the eddy current model in that the 8-Hz data show that the peak in $\chi_H''(T)$ is enhanced over that which is seen in the $\chi_0''(T)$ data. The 77-Hz data ($f>f_c$) show a peak in $\chi_H''(T)$ but none in $\chi_0''(T)$, a feature not permitted by the eddy current model. If a peak in $V_R(H,T)\approx\chi_H''(T)$ only occurs at frequencies less than f_c, then f_c (160 Oe) must be greater than 77 Hz or a factor of 4 larger than deduced from the $\chi_0''(T)$ data. The observation that $\chi_H''(N\text{-}S)\approx\chi_0''(N\text{-}S)$ rules out magnetoresistive effects on a/δ leading to an increase in f_c in the presence of a dc magnetic field.

In the case of Nb, data shown in Fig. 11, reveal that a dc magnetic field not only changes the height of the loss peak but it also removes the structure observed in the loss peak for $H=0$. One again notes that $\chi_H''(N\text{-}S)\approx\chi_0''(N\text{-}S)$.

It seems reasonable to conclude that eddy current models apply in the case where one has nearly ideal magnetic behavior. In the case of significant magnetic hysteresis losses, such losses will dominate eddy current effects resulting from a/δ considerations. The peaks in the $\chi_H''(T)$ data would then be associated with minor hysteresis loops resulting from the superposition of h_{ac} on H. In the $\chi_0''(T)$ data, the peak would be due to major hysteresis loops associated with h_{ac} (assuming the residual field $=0$). Since the loss peak in $H\neq0$ occurs at a lower temperature than the one in $H=0$, increased hysteresis effects are expected. One is tempted to attribute the behavior of the double peak structure, observed with Nb, as a manifestation of an interplay between magnetic hysteresis effects and eddy current effects; however one cannot rule out possible surface inhomogeneities as being responsible for this structure.[35]

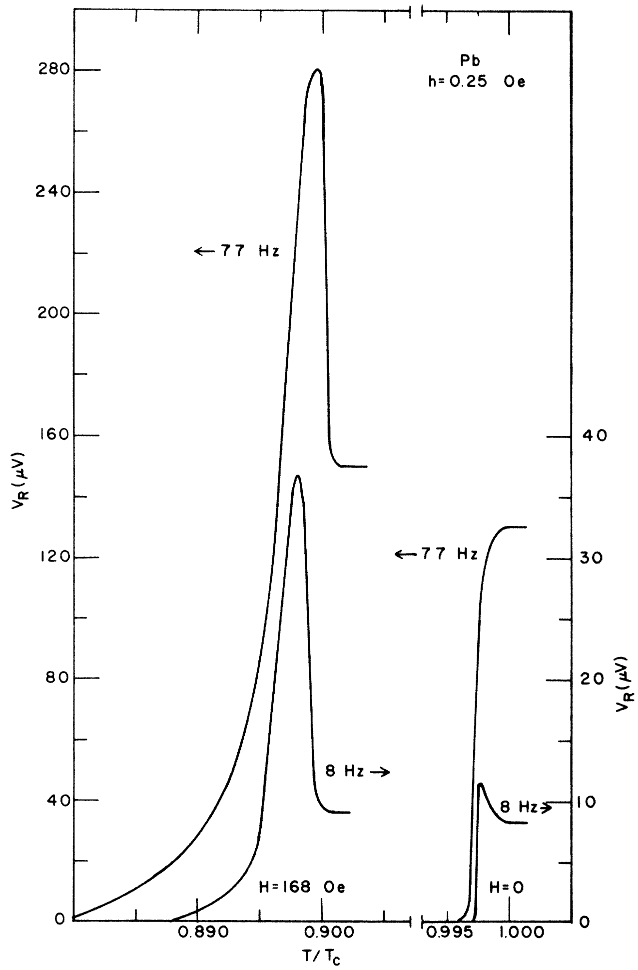

FIG. 10. V_R for the Pb cylinder as a function of the reduced temperature. Data are shown for two values of the applied dc magnetic field and for two values of the ac measuring frequency. The ac field strength was 0.25 Oe (rms). The ordinant is expressed in terms of the input voltage to the in-phase channel of the lock-in amplifier.

Recent studies of quasi-one-dimensional superconductors[36,37] have led to the development of yet another model to account for the observed dependences of the peak in $\chi_0''(T)$ upon both h_{ac} and f. This model assumes the sample to contain a multiply connected network of Josephson-type junctions. In this case[36] "for large h_{ac}, the magnetic flux passes the junction for every cycle and there occurs an effective resistance. This is the origin of χ''." Ishida and Mazaki[38] show that their weakly coupled loop model leads to expressions for χ' and χ'' which are

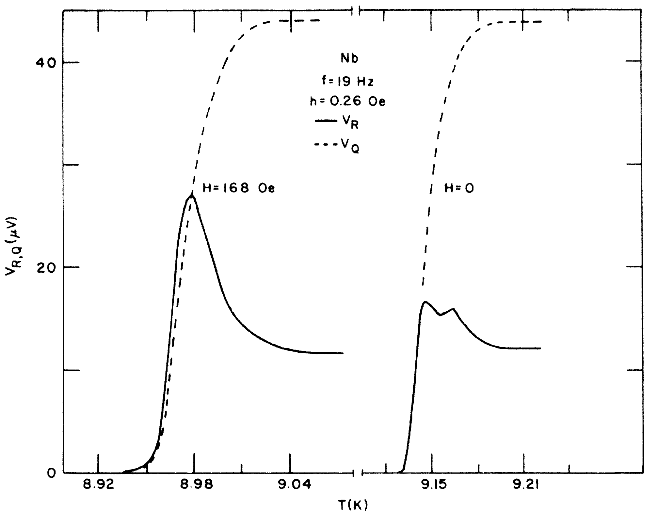

FIG. 11. V_R (solid curve) and V_Q (dashed curve) for the Nb cylinder as a function of temperature for two values of the applied dc magnetic field; the ac magnetic field was 0.26 Oe (rms). The ordinant is expressed in terms of the input voltages to the two channels of the lock-in amplifier.

equivalent to those derived from magnetic hysteresis models.[31,32,39]

Reflecting on the successes and shortcomings of the various models used to explain $\chi''(T)$ behavior one is inclined to agree with Rollins and Silcox[39] that the nature of the loss mechanism is not well understood and that perhaps some sort of relaxation mechanism is operative, a notion recently reintroduced by Campbell *et al.*[40]

V. CONCLUSIONS

Estimates of superconducting volumes based on the relative amplitudes of the "ac signal" for samples of comparable size and shape can result in gross overestimates of such volumes. Data of Figs. 1 and 4 indicate, in a quantitative manner, that such estimates can be in error by as much as two orders of magnitude. The technique of rendering a solid sample into a powder provides a useful test for determining if a diamagnetic signal observed with a solid sample is due to the presence of a superconducting minority phase. If one "grinds" a solid sample into a powder with particle dimensions comparable to, or greater than λ and observes an ac signal that is at least an order of magnitude smaller than the signal observed with the solid sample, he has reason to conclude that the larger diamagnetic signal associated with the solid sample was due to a minority phase present in the sample. However, if the powder produces an ac signal comparable to that of the solid sample, then one has to consider such things as the shape, distribution, and orientation of the particles making up the powder specimen. A small particle with a large demagnetization factor (e.g., $n \approx 0.9$) will, according to Eq. (2), yield a $\chi_0'(S)$ value corresponding to a much larger solid sample with $n \approx 0.1$. It is readily conceivable that a few judiciously located particles can mimic the ac signal of a much larger solid sample. If one "grinds" the sample into a powder with particles of dimensions less than λ, than a vanishingly small ac signal is to be expected even for a bona fide bulk superconductor. Thus, the simple technique of "powdering" a solid sample and remeasuring $\chi_0(T)$ will readily reveal if the "ac signal" is the result of a superconducting minority phase, but is of limited utility in the establishment of bulk superconductivity *per se*. The practice of setting $\chi_0'(S) = -1/4\pi$, for samples with nonzero demagnetization factors should be avoided.[37]

The observation that the ratio of the signal amplitude of a high-resistivity material, e.g., an alloy or compound, to that of a low-resistivity material, e.g., Pb, Sn, etc., is frequency dependent does not constitute a sufficient observation from which to conclude one is observing nonbulk superconductivity. Based on the recorder tracings shown in Fig. 2, one sees (a) the ratio of $V_Q(N\text{-}S) \approx \omega \Delta M'(N\text{-}S)$ for $Pb_{0.9}Bi_{0.1}$ to that of Pb at a given frequency is dependent on the frequency and (b) the $V_Q(N\text{-}S)$ values *per se* for $Pb_{0.9}Bi_{0.1}$ and Pb have different frequency dependences. This latter feature is a result of different frequency dependence inherent in $\Delta M'(N\text{-}S) = M'(N) - M'(S)$ for the samples involved, see Fig. 3. The discussion in Sec. IV B, based on the applicability of Eqs. (1) and (2), leads to the conclusion that the frequency-dependent ratio arises as a result of normal-state electrodynamics and need not, most likely does not, arise from different screening mechanisms in the superconducting states of the two samples.

The utility of using the presence or absence of an "extra-loss" peak in $\chi_0''(N\text{-}S)$ data to distinguish bulk from nonbulk or "filamentary" superconductors is seriously questioned by the data of Figs. 6, 7, and 8. These data strongly suggest that an extra-loss peak is ubiquitous to all N-S transitions provided one uses appropriate values for f and h_{ac}. From this result, one may conclude that the extra-loss peak is not a suitable observation upon which to base any conclusion with regard to bulk versus nonbulk superconductivity. Eddy current models[4,5,7,34] are capable of accounting for several facets of the zero-field ac magnetic-susceptibility data, i.e., $\chi_0'(T)$ and $\chi_0''(T)$, but appear incapable[3,7] of explaining frequency and h_{ac} effects on the loss peaks in $\chi_0''(N\text{-}S)$ and $\chi_H''(N\text{-}S)$, see Figs. 8—11. It appears that magnetic hysteresis effects must be playing a role; however, the mechanism or mechanisms responsible for these effects remains undefined.

There exists considerable confusion in the literature as to the significance of data obtained by magnetic induction techniques. Palstra *et al.*[41] in their discussion of bulk superconductivity in UPt, state that their sample showed nearly a full Meissner effect and then showed an irreversible magnetization curve displaying nearly 100% trapped flux. They considered the determination of the initial slope of the virgin magnetization curve as preferred data over that of ac susceptibility data. Why this should be so is not clear as both types of measurements reflect the "perfect shielding" aspects of the sample and one cannot deduce the Meissner effect or volume percentage that is superconducting from either set of data. The ring, sample *B* of Fig. 1, should have the "same" initial slope of the virgin magnetization curve as that of the solid cylinder. One also finds the use of the phrase "complete Meissner effect" or "Meissner effect" to describe perfect shielding associated with superconducting loops;[36,37] a practice which clearly should be avoided.

ACKNOWLEDGMENTS

The author gratefully acknowledges the experimental assistance of M. Colgan and G. Tohman and is indebted to J. Reimeika and B. Das for the $BaPbBiO_3$ and V_3Ga samples, respectively. This work was supported by U.S. Office of Naval Research (ONR) Contract No. N00014-82-K-0503.

[1]R. W. McCallum, W. A. Karlsbach, T. S. Radhakrishnan, F. Pobell, R. N. Shelton, and P. Klavins, Solid State Commun. **42**, 819 (1982); W. A. Karlsbach, R. W. McCallum, U. Poppe, F. Pobell, R. N. Shelton, and P. Klavins, in *Superconductivity in d- and f-band Metals*, edited by W. Buckel and W. Weber (Kernforschungszentrum Karlsruhe, Karlsruhe, 1982), p. 185.

[2]A. F. Khoder, Phys. Lett. **A94**, 378 (1983).

[3]R. A. Hein and R. L. Falge, Jr., Phys. Rev. **123**, 407 (1961).

[4]E. Maxwell and M. Strongin, Phys. Rev. **10**, 212 (1963); M. Strongin and E. Maxwell, Phys. Lett. **6**, 49 (1963).

[5]W. D. Gregory, in *The Science and Technology of Superconductivity*, edited by W. D. Gregory, W. N. Matthews, Jr., and E. Edelsack (Plenum, New York, 1973), Vol. II, p. 226.

[6]W. Meissner and R. Ochsenfeld, Naturwissenchaften **21**, 787 (1933).

[7]D. Shoenberg, Cambridge Philos. Soc. Proc. **33**, 559 (1937).

[8](a) C. W. Chu, S. Z. Huang, C. H. Lin, R. L. Meng, M. K. Wu, and P. H. Schmidt, Phys. Rev. Lett. **46**, 276 (1981). (b) C. W. Chu, S. Huang, and A. W. Sleight, Solid State Commun. **18**, 977 (1976).

[9]Y. Oda, H. Takenaka, H. Nagano, and I. Nakada, Solid State Commun. **32**, 659 (1979).

[10]M. Ribault, G. Benedek, D. Jerome, and K. Bechgaard, J. Phys. (Paris) Lett. **141**, 397 (1980).

[11]Y. J. Qian, J. O. Zheng, B. K. Sarma, H. O. Yang, J. B. Ketterson, and J. E. Hillard, J. Low. Temp. Phys. **49**, 279 (1982).

[12]Y. Oda and H. Nagano Solid State Commun. **35**, 631 (1980); Y. Oda, A. Sumiyama, and H. Nagano, Jpn. J. Appl. Phys. **22**, 464 (1983).

[13]W. Lieke, U. Rauchschwalbe, C. B. Bredl, F. Steglich, J. Aarts, and F. R. de Boer, J. Appl. Phys. **53**, 2111 (1982).

[14]M. S. Torikachvili, M. B. Maple, R. P. Guertin, and S. Foner, J. Low. Temp. Phys. **49**, 573 (1982). See newer negative results, in M. Decroux, S. E. Lambert, M. S. Torikachvili, M. B. Maple, R. P. Guertin, L. D. Woolf and R. Baillif, Phys. Rev. Lett. **52**, 1563 (1984); M. Decroux, S. E. Lambert, M. B. Maple, L. D. Woolf, R. Baillif, and O. Fischer, J. Low Temp. Phys. **60**, 149 (1985).

[15]R. P. Hudson, Phys. Rev. **29**, 283 (1950).

[16]H. R. Ott, H. Rudigier, Z. Fisk, and J. L. Smith, Phys. Rev. Lett. **50**, 1595 (1983).

[17]D. W. Harrison, K. C. Lim, J. D. Thompson, C. Y. Huang, P. D. Hambourger, and H. L. Luo, Phys. Rev. Lett. **46**, 280 (1981).

[18]R. L. Meng, T. H. Lin, M. K. Wu, C. W. Chu, and S. Z. Huang, J. Low Temp. Phys. **48**, 383 (1982).

[19]U. Rauschschwalbe, W. Lieke, C. D. Bredl, F. Steglich, J. Aarts, K. M. Martini, and A. C. Mota, Phys. Rev. Lett. **49**, 1448 (1982); F. Steglich, J. Aarts, C. D. Bredl, W. Lieke, D. Meschede, W. Franz, and H. Schafer, Phys. Rev. Lett. **43**, 1892 (1979).

[20]H. Schwenk, H. Andres, and F. Wudl, Phys. Rev. B **27**, 5846 (1983). K. Andres, F. Wudl, D. B. McWhan, G. A. Thomas, D. Nalewajek, and A. L. Stevens, Phys. Rev. Lett. **45**, 1449 (1980).

[21](a) B. N. Das, J. E. Cox, and R. W. Huber, U.S. Naval Research Laboratory (Washington, D.C. 20375) Report No. 7907, 1975 (unpublished); (b) B. N. Das, J. E. Cox, R. W. Huber, and R. A. Meussner, Metall. Trans. A **8A**, 541 (1977).

[22]E. Maxwell, Rev. Sci. Instrum. **36**, 533 (1965).

[23]R. A. Hein and P. H. E. Meijer, Phys. Rev. **179**, 497 (1969); The temperature-sweep method does not reveal the same details as does the swept magnetic field data of Ref. 3.

[24]M. H. Van Maaren, Phys. Lett. **29A**, 293 (1969); S. Ohshima, T. Wakiyama, T. Goto, and Y. Syona, Jpn. J. Appl. Phys. **22**, 264 (1983); J. L. Smith, Z. Fisk, J. O. Willis, B. Batlogg, and H. Ott, J. Appl. Phys. **55**, 1996 (1984).

[25]There exist numerous reports by workers concerned with ac losses; see M. A. R. Leblanc and J. P. Lorrain, Cyrogenics **24**, 143 (1984); and the following references, i.e., Refs. 26—35.

[26]M. Strongin, E. Maxwell, and T. B. Reed, Rev. Mod. Phys. **36**, 164 (1964).

[27]L. D. Landau and E. M. Lifshitz, *Electrodynamics of Continuous Media* (Pergamon, New York, 1960), p. 194.

[28]H. Sample, J. A. Gerber, L. J. Neoringer, and L. A. Kauffman, Phys. Lett. **33A**, 119 (1970). [This belief did not prevent the authors from using the magnitude of $\chi_0(N\text{-}S)$ to estimate that 50% or more of the sample was superconducting].

[29]M. Strongin, A. Paskin, D. G. Schweitzer, O. F. Kammerer, and P. P. Craig, Phys. Rev. Lett. **12**, 442 (1964); B. Bertman and M. Strongin, Phys. Rev. **147**, 268 (1966).

[30]A. Paskin, M. Strongin, P. P. Craig, and D. G. Schweitzer, Phys. Rev. **137**, A1816 (1965).

[31]H. J. Fink, Phys. Rev. Lett. **16**, 447 (1966); Phys. Rev. **161**, 417 (1967).

[32]C. A. M. van der Klein, J. D. Elen, R. Worf, and D. de Klerk, Physica **49**, 98 (1970).

[33]P. R. Doidge, Kwan Sik-Hung, and D. R. Tilley, Philos. Mag. **13**, 795 (1966).

[34]G. D. Cody and R. E. Miller, Phys. Rev. **173**, 481 (1968).

[35]T. Ishida and H. Mazaki, Phys. Rev. B **20**, 131 (1979).

[36]Y. Oda, H. Takenaka, H. Nagano, and I. Nakada, Solid State Commun. **35**, 887 (1980).

[37]T. Ishida, K. Kanoda, H. Mazaki, and I. Nakada, Phys. Rev. B **29**, 1183 (1984).

[38]T. Ishida and H. Mazaki, J. Appl. Phys. **52**, 6798 (1981).

[39]R. W. Rollins and J. Silcox, Phys. Rev. **155**, 404 (1967).

[40]S. A. Campbell, J. B. Ketterson, and G. W. Crabtree, Rev. Sci. Instrum. **54**, 1191 (1983).

[41]T. T. M. Palstra, P. H. Kes, J. A. Mydosh, A. DeVisser, J. J. M. Franse, and A. Menovsky, Phys. Rev. B **30**, 2986 (1984).