Glasslike low-frequency ac response of ZrB_{12} and Nb single crystals in the surface superconducting state

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We report experimental studies of the low-frequency electrodynamics of ZrB_{12} and Nb single crystals. The ac susceptibility at frequencies 3–1000 Hz has been measured under a dc magnetic field H_0 applied parallel to the sample surface. In the surface superconducting state for several H_0 the real part of the ac magnetic susceptibility exhibits a logarithmic frequency dependence as for spin-glass systems. Kramers-Kronig analysis of the experimental data shows large losses at ultralow frequencies (<3 Hz). The wave function slope at the surface was found. The linear response of the order parameter to the ac excitation was extracted from the experimental data.

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I. INTRODUCTION

Nucleation of the superconducting phase in a thin surface sheath in a decreasing magnetic field parallel to the sample surface was predicted by Saint-James and de Gennes some years ago.[1] They showed that nucleation of the superconducting phase occurs in a magnetic field $H_0 < H_{c3} = 2.39\kappa H_c$, where H_c is the thermodynamic critical field and κ is the Ginzburg-Landau (GL) parameter. Experimental confirmations of this prediction were done in a number of publications.[2–6] It was found that in the surface superconducting state (SSS) at low frequencies, the superconducting compounds are lossy materials. However, the losses in the SSS, which exceed the losses of the normal state, show a peak at external fields H_0, when $H_{c2} < H_0 < H_{c3}$. This peak exists even at frequencies of a few hertz. In general, the response is nonlinear and frequency dependent. The critical-state model was found adequate for a description of the experimental data.[5]

In the last few years the SSS has attracted renewed interest from various directions.[7–10,14,15,11] The stochastic resonance phenomena in the SSS for Nb single crystal were observed in the nonlinear low-frequency response to ac fields.[7] In Ref. 8 it was assumed that at $H_0 = H_{c3}$ the sample surface consists of many disconnected superconducting clusters and subsequently the percolation transition takes place at $H_{c3}^c = 0.81H_{c3}$. The paramagnetic Meissner effect is also related to the SSS.[9] Voltage noise and surface current fluctuations in Nb in the SSS have been investigated.[10] Surface superconducting states were detected also in single crystals of MgB_2 (Ref. 14) and ZrB_{12} (Ref. 15). A new theoretical approach, based on a generalized form of the GL functional, was developed in Ref. 11. Surface superconductivity in three dimensions was studied theoretically in Refs. 12 and 13. It was found[5,15] that, in general, the wave form of the surface current in an ac magnetic field has a nonsinusoidal character. The latter can be described by a simple phenomenological relaxation equation for transitions between metastable states.[15] The relaxation time in this equation depends on the deviation from equilibrium and increases with decreasing of the excitation frequency.[15] In spite of the extensive study, the origin of low-frequency losses in SSS under weak ac fields is not clear yet. The critical-state model implies that if the amplitude of the ac field is smaller than some critical value, the losses are absent. Indeed, the experimental results for Pb-2% In alloy confirm this prediction.[5] On the other hand, the observed response[8] for an excitation amplitude of 0.01 Oe (which is, considerably smaller than that used in Ref. 5) shows losses in SSS of Nb at 10 Hz. Our measurement on Nb single crystal at 733 Hz (Fig. 2) also has shown that the out-of-phase part of the ac susceptibility, χ'', is independent of the amplitude and is finite at low amplitude values. We consider this experimental result as evidence of the linear origin of the losses in SSS, and thus the critical-state model cannot be used for an adequate description of the ac response.

In this paper we present a detailed experimental study of the linear low-frequency response of ZrB_{12} ($T_c = 6.06$ K, $\kappa \approx 0.75$) and Nb ($T_c = 9.2$ K, $\kappa \approx 1.5$) single crystals in the SSS for frequencies $3 \leqslant \omega/2\pi \leqslant 1000$ Hz. We show that already at 3 Hz the ac susceptibility does not equal $\partial\widehat{M}/\partial H_0$, where $\widehat{M}$ is the dc equilibrium magnetization. For some dc magnetic fields $\chi' \propto \ln \omega$, which resembles spin-glass systems.[17] The Kramers-Kronig analysis of our experimental data predicts that for several dc magnetic fields huge loss peak should exist at very low frequencies. It is believed that the observed response presents the average value over many clusters, each of which is governed by a second-order differential equation, with individual relaxation parameters. This is unlike a spin-glass system where the first-order differential equation can be used.[18] The observed ratio of H_{c3}/H_{c2} for our crystals differs from the predicted value: 1.69. It is believed that this is due to the nonzero slope of the wave function at the surface, $b = -(1/\Psi)(\partial\Psi/\partial x)$. The increased ratio of H_{c3}/H_{c2} shows that the sign of b is unexpectedly negative.[19]

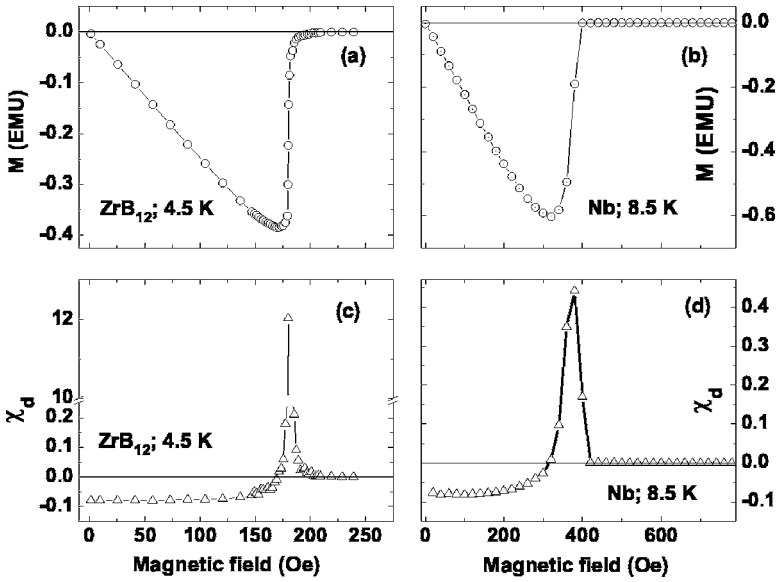

FIG. 1. (a), (b) Magnetization curves of ZrB_{12} and Nb samples after ZFC. (c), (d) Differential dc susceptibility $\chi_d \equiv 1/V \times [dM/dH(H_0)]$ of ZrB_{12} and Nb samples, numerical calculations.

The order parameter Ψ in the SSS has the form

$$\Psi(x,y) = f(x)\exp(iKy), \tag{1}$$

where $f(x)$ equals zero inside the bulk of the sample (the x axis is assumed normal to the sample surface, H_0 parallel to the z axis) and K is some as yet undetermined constant. In the general case for any dc magnetic field there is a band of possible values for K for which the solution of the GL equations can be found. These solutions describe the metastable surface states with nonzero total surface current, and only the solution with zero current corresponds to the equilibrium state. In this state the dc magnetic moment equals zero for $H > H_{c2}$. On the other hand, the nonzero ac response in SSS clearly shows that the ac moment is differ from zero in the ac field; thus, the system does not remain in equilibrium states with equilibrium K's. In the linear approximation,

$$K(t) = K_0 + \mathrm{Re}[G(\omega,H_0)h(\omega)\exp(-i\omega t)], \tag{2}$$

where the external magnetic field is

$$H(t) = H_0 + \mathrm{Re}[h(\omega)\exp(-i\omega t)]$$

and K_0 is equilibrium K in dc field H_0. The function $G(\omega,H_0)$ characterizes the response of the order parameter to an ac field and is determined in this paper.

II. EXPERIMENTAL DETAILS

The measurements were carried out on ZrB_{12} and Nb single crystals, which were grown in the Institute for Problems of Materials Science NAS, Ukraine, and in the Institute of Solid State Physics RAS, Russia, respectively. The dimensions of the crystals are $10.3 \times 3.2 \times 1.2$ mm^3 for ZrB_{12} and $10 \times 3 \times 1$ mm^3 for Nb. The details of the sample preparation and their magnetic characteristics were published previously.[7,20] The magnetization curves were measured using a superconducting quantum interference device (SQUID) magnetometer. Both dc and ac measurements were performed parallel to the longest side of the sample dc magnetic field. ac susceptibility, in-phase, χ', and out-of-phase, χ'', components were measured using the pickup coils method[5,21] with dc and ac fields parallel to each other. The sample was inserted into one of a balanced pair coils. The unbalanced signal as a function of the external parameters—temperature, dc magnetic field, frequency, and amplitude of excitation—was measured by a lock-in amplifier. The experiment was carried out as follows. The crystal was cooled down at zero magnetic field (ZFC). Then the magnetic field was applied. The amplitude and phase of the unbalanced signal were measured in a given magnetic field, including zero field, at all frequencies. The amplitude of excitation was 0.01–0.5 Oe. We have supposed that in zero dc magnetic field the ac crystal susceptibility is equal to the dc susceptibility in the Meissner state with negligible losses. It permits us to find an absolute value of the in-phase and out-of-phase components of the ac magnetic susceptibility for all applied fields and frequencies. A "homemade" measurement cell of the experimental setup was adapted to a commercial SQUID magnetometer. The block diagram of the experimental setup has been published elsewhere.[15]

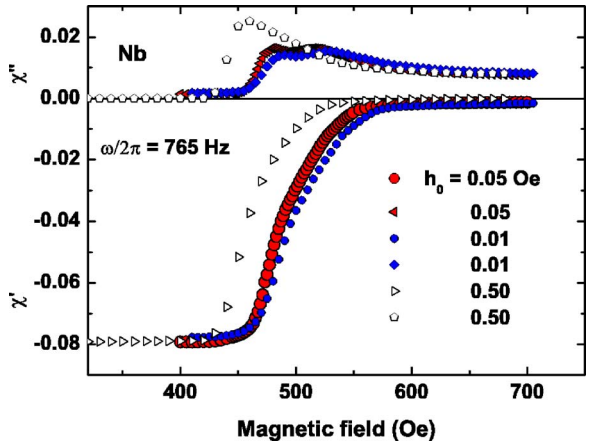

FIG. 2. (Color online) Magnetic field dependences of χ' and χ'' for Nb at $T = 8.5$ K at different amplitudes of excitation, h_0.

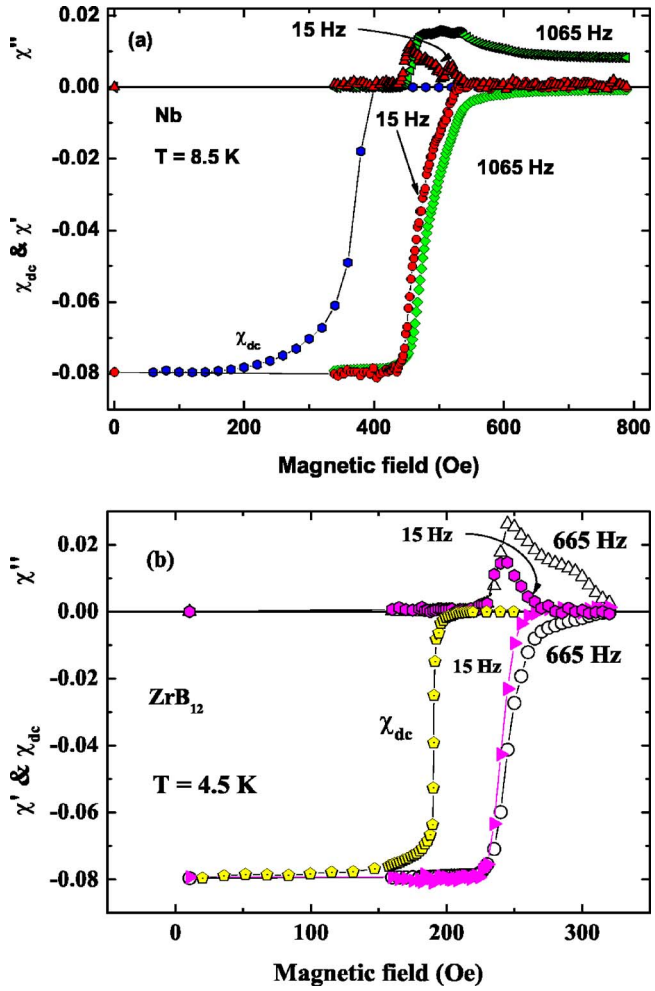

FIG. 3. (Color online) (a) Magnetic field dependences of χ', χ'', and $\chi_{dc}=1/V\times M/H_0$ for Nb at $T=8.5$ K. (b) Magnetic field dependences of χ', χ'', and $\chi_{dc}=1/V\times M/H_0$ for ZrB_{12} at $T=4.5$ K.

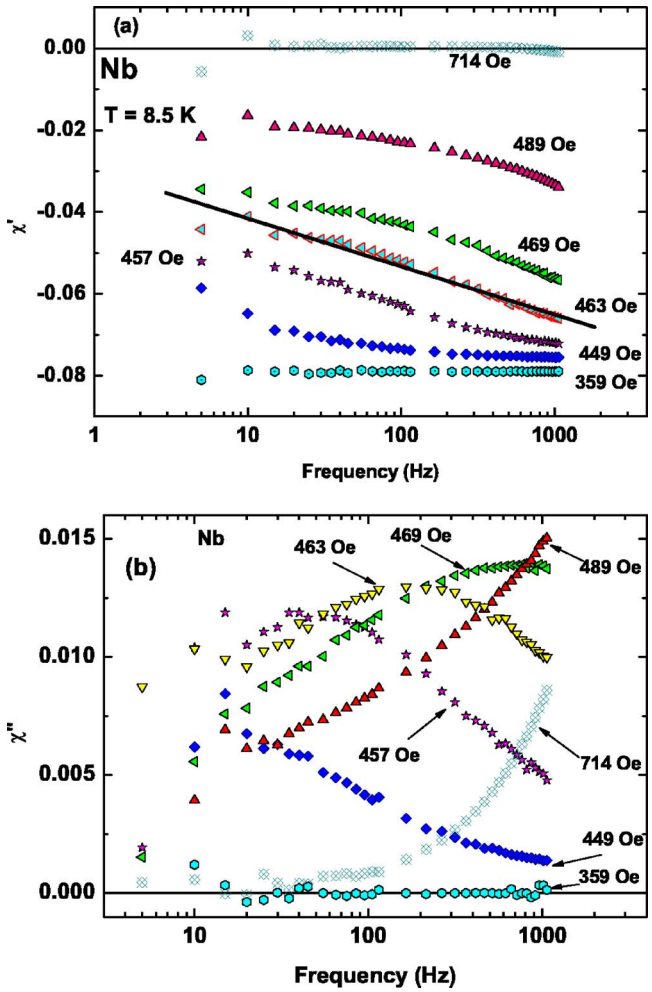

FIG. 4. (Color online) Frequency dependence of the ac susceptibility for Nb at $T=8.5$ K at dc magnetic field $H_0>H_{c2}$. (a) $\chi'(\omega)$ and (b) $\chi''(\omega)$.

For the Fourier component of the magnetization $m(t)=\mathrm{Re}[m(\omega,H_0,h(\omega))\exp(-i\omega t)]$ in the linear approximation one can write

$$m(\omega,H_0,h(\omega))=\chi_n[h(\omega)+4\pi J_s(\omega,H_0,h(\omega))/c]+4\pi\chi_s J_s(\omega,H_0,h(\omega))/c, \quad (3)$$

where χ_n is the susceptibility of the normal core of the sample and $J_s(\omega,H_0,h(\omega))$ is the Fourier component of the surface supercurrent, $J_s(t)=\mathrm{Re}[J_s(\omega,H_0,h(\omega))]\exp(-i\omega t)$. This equation takes into account the magnetic moment of the normal core of the bulk and the magnetic moment of the surface supercurrent.[5] With Eq. (3) we can find $J_s(\omega,H_0,h(\omega))$ and the surface susceptibility defined as $\chi_s(\omega,H_0)\equiv J_s(\omega,H_0,h(\omega))/h(\omega)$. The measured susceptibility $\chi(\omega,H_0)\equiv m(\omega,H_0,h(\omega))/h(\omega)$ and surface susceptibility χ_s are connected to each other as follows:

$$\chi_s(\omega,H_0)=[\chi(\omega,H_0)-\chi(\omega,H_n)]/[1+4\pi\chi(\omega,H_n)], \quad (4)$$

where $H_n>H_{c3}$ is the magnetic field at which the sample is in the normal state. These quantities (J_s and χ_s) characterize the response of the surface current and eliminate the contribution of the normal core in the bulk and the small unbalanced signal of the empty coils.

III. EXPERIMENTAL RESULTS

Figures 1(a) and 1(b) show the virgin magnetization curves of crystals ZrB_{12} and Nb. One can define H_{c2} for both samples using the Abrikosov conclusion that near H_{c2} the sample magnetization $M\propto(H_{c2}-H_0)$.[16] The field dependence of the differential susceptibility $\chi_d\equiv 1/V\times dM/dH_0$, where V is the sample volume, was calculated numerically on the basis of the experimental data. Figures 1(c) and 1(d) show $\chi_d(H_0)$ for ZrB_{12} and Nb samples, respectively. In magnetic fields above H_{c2} the magnetic moment M and differential dc magnetic susceptibility become negligibly small.

The field dependences of the ac susceptibility χ' and χ'' of Nb single crystal at different excitation amplitudes (h_0) are presented in Fig. 2. The data show clearly that at low amplitudes χ' and χ'' are almost independent of the h_0. Similar results were obtained for the ZrB_{12} crystal. One can consider that at $h_0\leqslant 0.05$ Oe the observed response in both Nb and ZrB_{12} crystals has a linear origin.

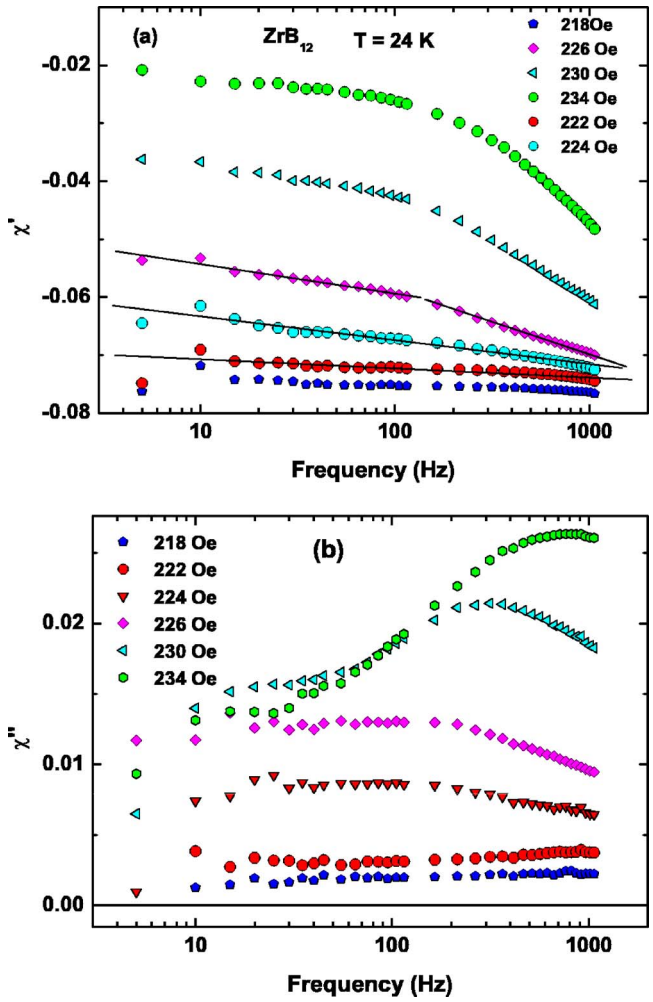

FIG. 5. (Color online) Frequency dependence of the ac susceptibility for ZrB_{12} at $T=8.5$ K at dc magnetic field $H_0 > H_{c2}$. (a) $\chi'(\omega)$ and (b) $\chi''(\omega)$.

Figures 5(a) and 5(b) show the χ' and χ'' as a function of the dc field (H_0) of Nb and ZrB_{12}, respectively. For both crystals, for $H_0 > H_{c2}$ both M and dM/dH are zero (Fig. 4) while the large ac susceptibility is observed (Fig. 4). We observed this even at $\omega/2\pi=3$ Hz (the lowest frequency in our setup). The out-of-phase component χ'' has a broad maximum for $H_0 > H_{c2}$. It is clear that the observed ac response is not connected with static magnetization because $M=0$ in these dc fields. Moreover, dM/dH exhibits large maximum near H_{c2} but the ac signal does not show any peculiarity at these fields.

In the normal state, for $H_0 > H_{c3}$, the losses for Nb are greater than for ZrB_{12} [see Fig. 2(a) for 1065 Hz], because the normal-state conductivity of Nb is larger than that of ZrB_{12}. The out-of-phase susceptibility χ'' is proportional to ω as expected for the normal skin effect in the limit $\delta \gg d$, where δ is the skin depth and d is the sample thickness.[22] For both crystals, when $H_0 > H_{c3}$, χ'' is field independent. However, in SSS (see Fig. 3), the losses for Nb are lower than for ZrB_{12}. Both the in- and out-of-phase components of the ac susceptibility, $\chi(\omega, H_0)$, do not show any clear peculiarities as the dc magnetic field passes through H_{c3}. Therefore, it is difficult to extract the H_{c3} value from the experimental data. A more sensitive measurement of H_{c3} can be done by using the $\chi_s(\omega, H_0)$ curves (see below).

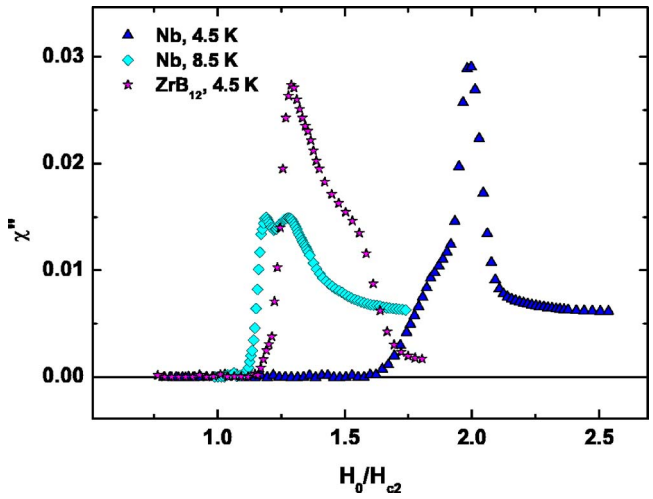

FIG. 6. (Color online) The field dependence of χ'' for Nb at $T=4.5$ K and 8.5 K and for ZrB_{12} at $T=4.5$ K (where $\omega/2\pi=765$ Hz).

Figures 4 and 5 demonstrate the frequency dependence of χ' and χ'' for both Nb and ZrB_{12} crystals at dc magnetic field above H_{c2}. The character of these curves depends on the field. Thus χ' is a concave function of ω at fields close to H_{c2} and is a convex function close to H_{c3}. For some fields, $\chi' \propto \ln \omega$ for both samples. Such a logarithmical dependence is typical of spin glasses which are completely different physical systems.[17] Figure 6 presents χ'' as a function of the reduced magnetic field, H_0/H_{c2}, for two frequencies and temperatures for Nb and ZrB_{12} crystals. The H_{c3}/H_{c2} ratio is temperature dependent as previously reported for Nb (Ref. 23).

IV. THEORETICAL BACKGROUND

For interpretation of the experimental data we used the numerical approach to the normalized stationary GL equations

$$-(i\nabla/\kappa + \vec{A})^2\Psi^2 + \Psi - |\Psi|^2\Psi = 0,$$

$$-\mathrm{curlcurl}\vec{A} = \vec{A}|\Psi|^2 + i/2\kappa(\Psi^* \nabla \Psi - \Psi \nabla \Psi^*), \qquad (5)$$

for an external magnetic field parallel to the sample surface. The order parameter Ψ is normalized with respect to its value at zero magnetic field, the distances with respect to the London penetration length λ, and the vector potential $\vec{A}$ with respect to $\sqrt{2}H_c\lambda$, where H_c is the thermodynamic critical field. Assuming that the order parameter has the form of Eq. (1) with the as yet undetermined parameter K, we see that K is the integral constant of these equations if the sample thickness considerably exceeds the coherence length and the superconductor is homogeneous. The relaxation time in the time-dependent version of GL theory is of the order of $10^{-11}-10^{-13}$ s and we can use the stationary version of the

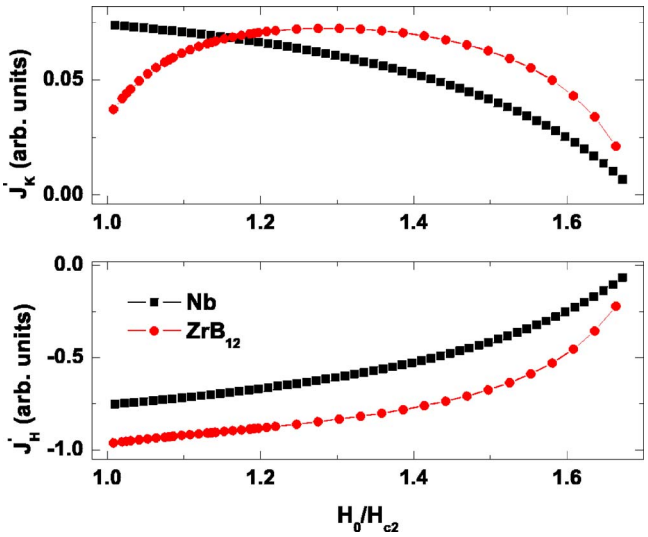

FIG. 7. (Color online) The derivatives of the surface current, J'_H and J'_K, versus reduced magnetic field for Nb and ZrB_{12} (see text).

GL theory, Eq. (5), for the ac experiments. Equations (5) with proper boundary conditions and the requirement that the order parameter differ from zero only near the surface have the solution for a whole band of K values. But only one value of K corresponds to the situation in which the total surface current equals zero and this K describes the equilibrium state with the minimal free energy. These nonlinear equilibrium surface solutions have been discussed in Ref. 24. The ac response for superconductors in fields $H_{c2}<H_0<H_{c3}$ differs from the normal one. This means that in an ac field the superconductor is in a nonequilibrium state with finite surface current and nonequilibrium K. Generally speaking, the total surface current depends on both the instant values of external magnetic field $H(t)$ and $K(t)$, thus $J_s(t)=J_s(H(t),K(t))$. If a small ac magnetic field $h(t)$ is superimposed upon a dc field H_0, the amplitude of the surface current in the linear approximation is

$$J_s(\omega,H_0,h(\omega)) = \frac{\partial J_s(H_0,K_0)}{\partial H_0}h(\omega) + \frac{\partial J_s(H_0,K_0)}{\partial K_0}G(\omega,H_0)h(\omega), \qquad (6)$$

where K_0 is the equilibrium value of K in a dc magnetic field H_0 and $G(\omega,H_0)$ describes the linear response of K to an ac field, Eq. (2). The partial derivatives in Eq. (6) ($J'_K \equiv \partial J_s(H_0,K_0)/\partial K_0$ and $J'_H \equiv \partial J_s(H_0,K_0)/\partial H_0$) can be calculated numerically and in Fig. 7 we show the results for Nb [GL parameter $\kappa=1.5$ (Ref. 23)] and ZrB_{12} [$\kappa=0.75$ (Ref. 20)].

Equation (6) describes the linear response of the surface current to an external ac magnetic field. If K does not change during an ac cycle, we see from Fig. 7 that only a smooth decrease of the surface current without any losses should be observed, as the dc magnetic field increases. The critical current model assumes that for $h(\omega)<h_c$, where h_c is some critical ac field, the surface current follows the external magnetic field without any delay, Im $G(\omega,H_0)=0$, and therefore surface losses are absent.

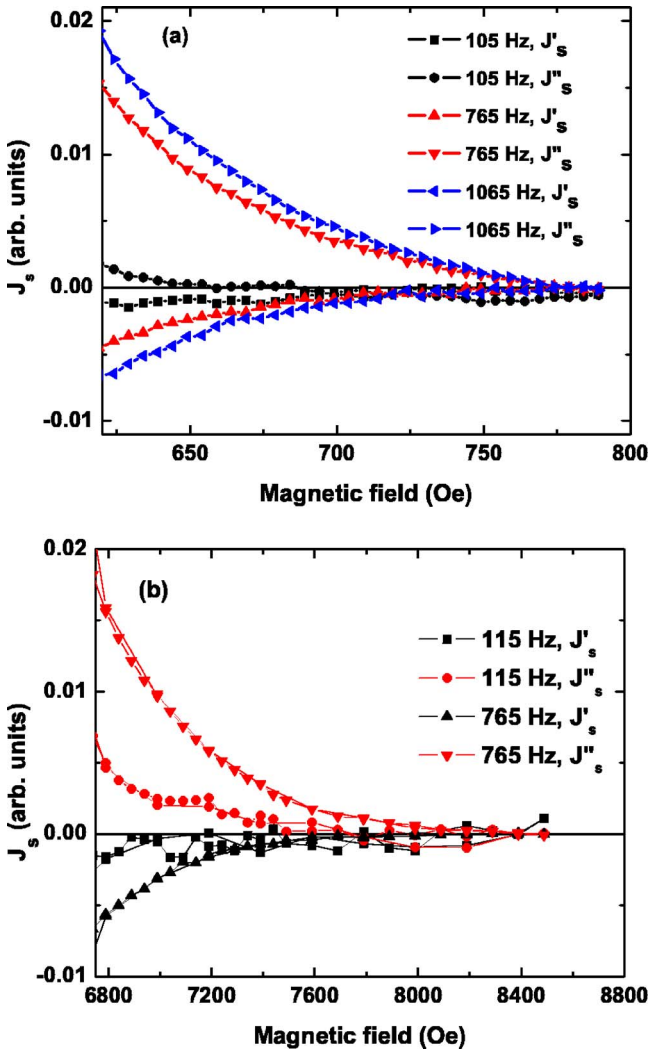

FIG. 8. (Color online) The field dependence of the real, J'_s, and imaginary, J''_s, parts of surface current at different frequencies for Nb at $T=8.5$ K, $H_{c2}=400$ Oe (a), and 4.5 K, $H_{c2}=3200$ Oe (b).

V. DISCUSSION

It is clear that the experimental data show the existence of SSS in our crystals. Usually H_{c3} is defined by the onset of the surface screening—i.e., by the appearance of the deviation of the ac response from its normal value, as the dc field decreases from some large value. Because in the SSS the ac response is frequency dependent, using low frequencies could give an underestimated value of H_{c3}. As an example, in Fig. 8(a) we show a set of data obtained for Nb sample at 8.5 K at frequencies 115 and 765 Hz. The imaginary part of surface current is more sensitive for H_{c3} determination (obviously, the experimental setup sensitivity is higher at higher frequencies). The data at a frequency of about 100 Hz yield $H_{c3}\cong 680$ Oe, while at higher frequencies ($\approx$700 Hz) we obtain $H_{c3}\cong 760$ Oe, in the later case $H_{c3}/H_{c2}=1.9$. This value

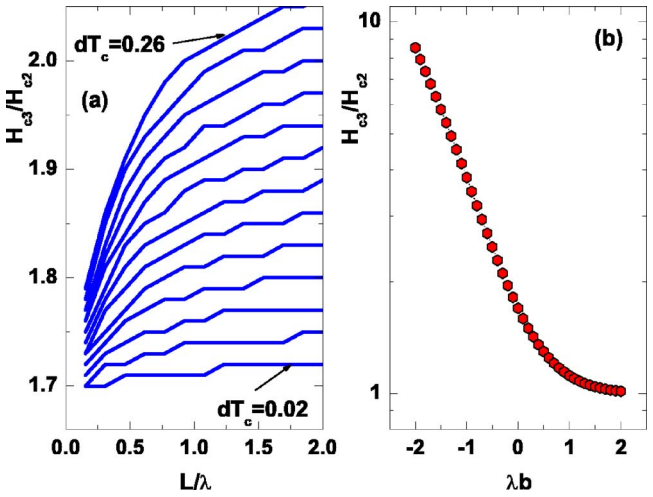

FIG. 9. (Color online) The H_{c3}/H_{c2} ratio for two cases: (a) T_c in the surface layer (of thickness L) is higher than in the bulk value [see Eq. (7)] and (b) the slope of the wave function, b, at the surface is different from zero ($\kappa=1.5$).

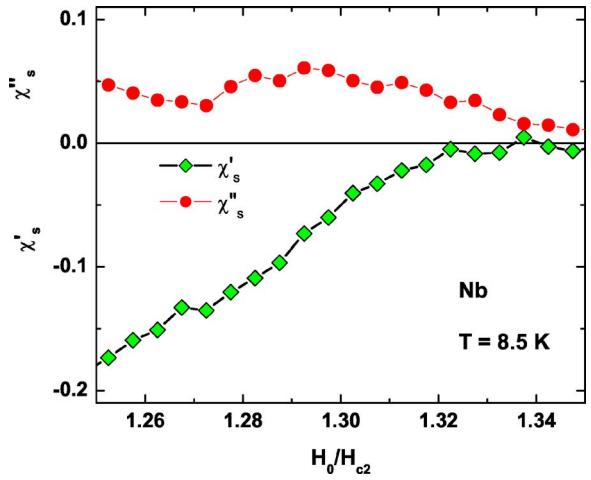

FIG. 10. (Color online) Real and imaginary parts of surface susceptibilities versus the dc magnetic field for Nb at 20 Hz and $T=8.5$ K near the point where the absolute values of both components are equal to each other ($H_0/H_{c2}\approx 1.3$).

is considerably larger than the value predicted 1.69 in Ref. 1. With decreasing temperature, the discrepancy between the theoretical prediction[1] and the experimental values increases and the ratio at $T=4.5$ K becomes $H_{c3}/H_{c2}=2.34$ [Fig. 8(b)]. The decrease of this ratio for temperatures in the vicinity of T_c was found in several experiments,[23,25] and it was associated with the decrease of T_c near the surface.[26] In the framework of the GL theory the H_{c3}/H_{c2} ratio can be changed only if either (i) T_c in the surface layer differs from the bulk value or (ii) the slope of the wave function at the surface $b=-(1/\Psi)(\partial\Psi/\partial x)$ differs from zero.[19,27] In Fig. 9 we show the calculated $H_{c3}/H_{c2}(\kappa=1.5)$ for these two cases.

The boundary condition for the first case is $b=0$ but the T_c values of a surface layer with thickness L increases as

$$[T_c(x)-T_c]/(T_c-T)=dT_c\exp(-x/L) \tag{7}$$

[Fig. 9(a)]. In the second one (when $dT_c=0$), we assume that $b\neq 0$ [Fig. 9(b)]. The ratio of $H_{c3}/H_{c2}\simeq 1.9$ at 8.5 K; this can be a result of either the enhanced T_c by 0.13 K ($dT_c=0.2$) at the surface layer with the thickness of $L/\lambda=0.71$ or by assuming $b=-0.15/\lambda$. Decreasing the temperature results in an increase of both the L/λ and H_{c3}/H_{c2} ratios. At $T=4.5$ K, $dT_c=0.028$, and from Fig. 9(a) one finds that the value of the H_{c3}/H_{c2} cannot exceed 1.75. Therefore, if the GL theory is applicable, then, at $T=4.5$ K, the growth of the H_{c3}/H_{c2} to 2.34 in Nb crystal is due to $b\neq 0$ and the absolute value of b increases with decreasing temperature.

The behavior of the ZrB_{12} crystal is similar. The H_{c3}/H_{c2} ratio increases from 1.5 at 5.5 K to 1.75 at 4.5 K. Since this ratio at 5.5 K is smaller than 1.69, it is possible that T_c of the surface layer is smaller than that of the bulk. Our calculation shows that this value (1.5) can be obtained, for example, if $dT_c=0.16$ and $L/\lambda=2$. For $T=4.5$ K the dimensionless dT_c is equal to 0.05 and the H_{c3}/H_{c2} ratio does not exceed 1.65. We notice that in order to explain the H_{c3}/H_{c2} ratio, one has to take into account the nonzero slope of the wave function at the surface.

In general, with decreasing the dc field from its maximal value, both the real and imaginary parts of the surface current appear simultaneously, but at the beginning the imaginary part increases faster than the real one. On the other hand, at H_{c2} a complete screening takes place and the absolute value of χ_s' reaches its maximal value $1/4\pi$, while $\chi_s''=0$. So at some dc magnetic field $|\chi_s'|$ will be equal to $|\chi_s''|$. This point was identified in Ref. 8 as the percolation transition from noncoherent SSS to the coherent one in the Nb sample. For our Nb crystal, at $T=8.5$ K and frequency 20 Hz it occurs at $H_0/H_{c3}\approx 0.68$, a value which is slightly smaller than the value 0.81 reported in Ref. 8. However, our data presented in Fig. 10 do not permit us to consider this point as a phase transition, due to a smooth maximum $|\chi_s''|$ at this field and the absence of any peculiarity in χ_s'.

The response of K to an ac field is described by the function $G(\omega,H_0)$ which can be found from Eq. (6). Figures 11(a) and 11(b) show $G(\omega,H_0)$ as a function of a reduced magnetic field at several frequencies. Generally, $G(\omega,H_0)$ depends on the frequency. Moreover, the real part of $G'(\omega,H_0)$ changes its sign with increasing dc field. The frequency dependence of $G'(\omega,H_0)$ of the Nb crystal for several magnetic fields near the loss peak, presented in Fig. 11, is shown in detail in Fig. 12(a). For some magnetic fields $G'(\omega,H_0)$ changes its sign as the frequency increases. The zero in the real part of the response function can be considered as the manifestation of the existence of unknown collective mode in the system. In Fig. 12(b) we show the frequency of this hypothetical surface mode as a function of the dc field for Nb at 8.5 K. The dynamics of K cannot be described by the first-order linear differential equation

$$\partial K/\partial t=-\nu[K-\gamma h(t)] \tag{8}$$

with some relaxation constant ν and $\gamma=-\partial J(H_0,K_0)/\partial H_0\big/\partial J(H_0,K_0)/\partial K_0>0$. Equation (8) gives the real part $G'(\omega,H_0)=\gamma\nu^2/(\nu^2+\omega^2)$, which does not change its sign with frequency. One could expect that the

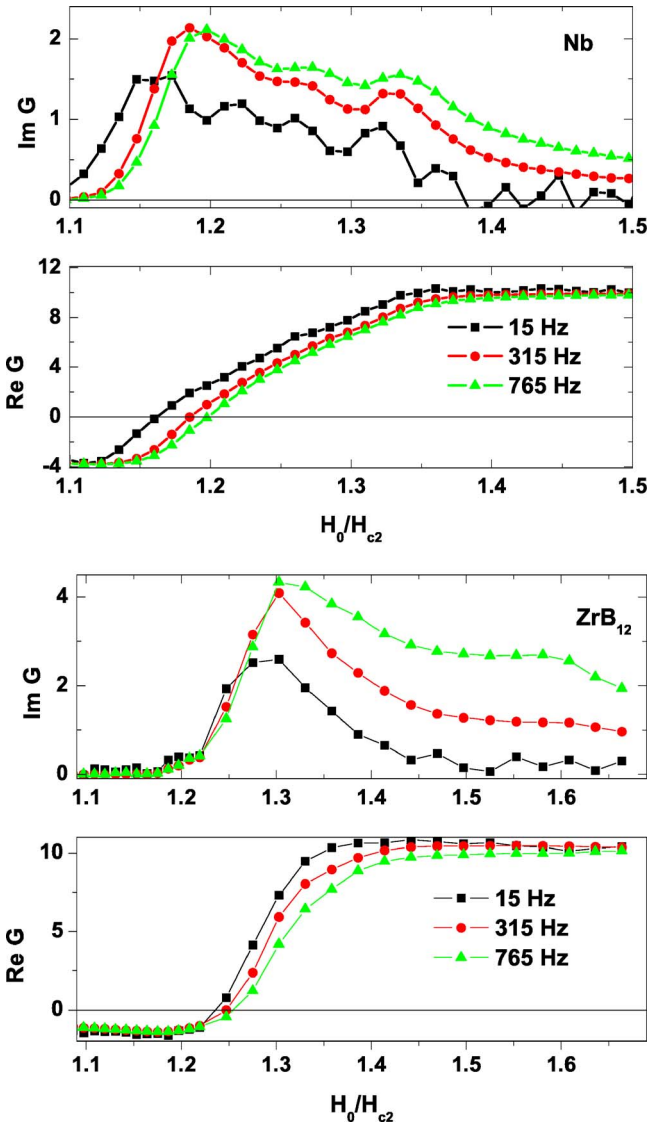

FIG. 11. (Color online) The response function $G(\omega,H_0)$ of Nb and ZrB_{12} samples (upper and lower panels, respectively) versus reduced magnetic field, H_0/H_{c2}.

second-order differential equation will give an adequate description of the observed response. But our experimental data show that $G'(\omega,H_0)$ cannot be obtained from a differential equation of comparatively low order.

The observed logarithmic frequency dependence of χ', Fig. 3(a) for some dc fields, resembles the response of a spin-glass system. But in spin-glass materials χ'' is a slow function of the frequency as compared to χ'. Our data show that the χ' and χ'' values both depend on the frequency. Probably, similar to the spin-glass systems, we have here a lot of clusters which are governed by the second-order differential equation; thus, the observed response is the average over all clusters. The second-order differential equation

$$\partial K/\partial t = -\nu(K - \gamma h(t)) - \beta\partial^2 K/\partial t^2, \tag{9}$$

with $\beta > 0$, has a response that changes its sign at $\omega_0^2 = \nu/\beta$. Assuming that ν/β for all clusters increases with the dc field,

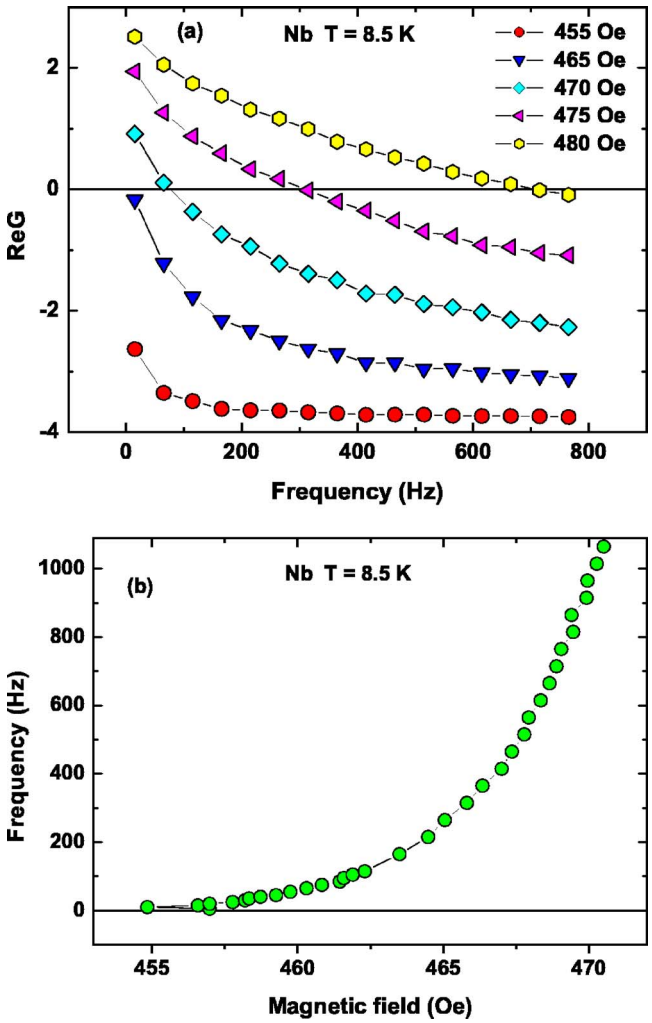

FIG. 12. (Color online) (a) Frequency dependence of G'' at different magnetic fields near the absorbtion maximum for Nb crystal at $T=8.5$ K. (b) Dispersion relation of the surface mode, $\omega_0(H_0) = \sqrt{(\nu/\beta)}$, for a Nb sample at $T=8.5$ K.

we find that the dc field value for which $G'(\omega,H_0)=0$ must increase with the frequency, as observed in Fig. 11. The $\sqrt{\nu/\beta}$ quantity in this case will be the frequency of the surface collective mode. The dispersion relation of this mode, $\omega_0(H_0)$, for Nb is shown in Fig. 12(b).

Further insight into the low-frequency response can be obtained from the Kramers-Kronig relation[22]

$$\chi_s'(\omega) - \chi_\infty = \frac{2}{\pi}\int_0^\infty \frac{\xi\chi_s''(\xi)d\xi}{\xi^2 - \omega^2}, \tag{10}$$

where $\chi_\infty = \chi_s(\infty)$. If we chose $\xi_0 \ll \omega \ll \xi_m$, then

$$\chi_s'(\omega) - \frac{2}{\pi}\int_{\xi_0}^{\xi_m} \frac{\xi\chi_s''(\xi)d\xi}{\xi^2 - \omega^2} = \chi_\infty + \frac{2}{\pi}\left(-\int_0^{\xi_0} \frac{\xi\chi''(\xi)d\xi}{\omega^2} + \int_{\xi_m}^\infty \frac{\xi\chi_s''(\xi)d\xi}{\xi^2}\right). \tag{11}$$

The left side of this equation can be extracted from the avail-

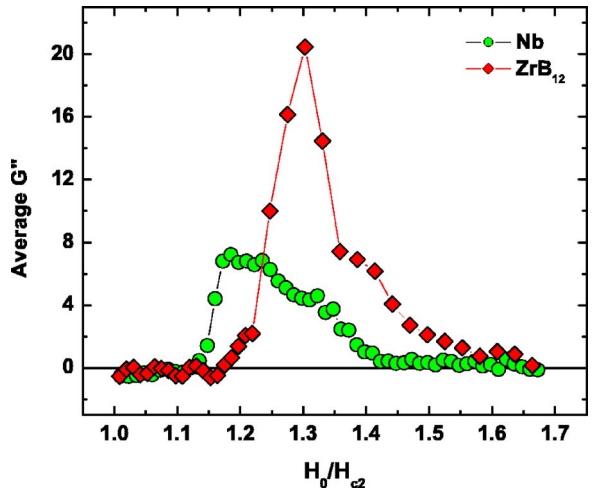

FIG. 13. (Color online) $\overline{G''(\omega,H_0)}$ over the frequency interval $0 \leqslant \omega/2\pi \leqslant 20$ Hz as a function of reduced magnetic field, H_0/H_{c2}, for Nb at 8.5 K and ZrB_{12} at 4.5 K.

able experimental data. By presenting it as a linear function of $1/\omega^2$ one can obtain the averaged imaginary part at low frequency, defined as

$$\overline{\chi_s''}(\xi_0) = 2\int_0^{\xi_0} \xi\chi_s''(\xi)d\xi/\xi_0^2. \tag{12}$$

In Fig. 13 we have shown the averaged imaginary part of $\overline{G''}(\xi_0)$ $(G'' \propto \chi_s'')$, obtained for Nb at $T=8.5$ K, using $\xi_0 = 2\pi 20$ s^{-1} and $\xi_m = 2\pi 1065$ s^{-1}. $\overline{G''}(\xi_0)$ in Fig. 12 is considerably larger than any value shown in Fig. 11. So the Kramers-Kronig relations predict the existence of a large loss peak at low frequencies. This prediction needs further experimental evidence.

The observed ac response of SSS has a very complex character. Partially, it can be the result of nonhomogeneity of the samples. The K parameter, as it was introduced by Eq. (1), corresponds to the wave function of the whole sample. In real samples, due to inhomogeneity, K is not the integral constant of the GL equations and the exact wave function is the superposition of states with different K. Those states can relax with different relaxation times as in spin-glass systems,[17] and therefore we observe such complicated responses. In addition, the dynamics of the surface state is not described by a first-order simple relaxation equation. As a result, we obtain the logarithmic frequency dependence of the real part of the susceptibility, as in spin-glass systems, but the imaginary part shows a maximum at some frequency.

VI. CONCLUSIONS

In this paper we have presented investigation of the linear ac susceptibility of Nb and ZrB_{12} single crystals in the surface superconducting state. Losses in this state have a linear origin, and the critical-state model for the surface current does not apply here. Similar to spin-glass systems (where finite losses at considerably low frequencies exist), the real part of susceptibility exhibits a logarithmic frequency dependence. But the out-of-phase component has a frequency dispersion. This dispersion in SSS differs from that of the spin-glass systems. We assume that the sample surface presents a lot of superconducting clusters, which are governed by a second-order differential equation, and the observed response is an average over these clusters. The Kramers-Kronig analysis of experimental data reveals huge absorption peak at low frequencies. The response of K to the ac magnetic field defined by Eq. (1), $G(\omega,H_0)$, has been measured.

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