Nonlinear interaction of sound waves in metals

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The interaction in metals of variously polarized sound waves has been investigated. An amplification type effect was detected in gallium in the normal state under momentum nonlinearity conditions. A "step" type nonlinearity and suppression of the effect of weak superconducting ordering on the sound attenuation was observed in superconducting gallium near the transition temperature T_c. The nonlinear attenuation of sound in gallium and aluminum was also investigated far from T_c.

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The introduction of an intense sound wave into a pure metal results in a distortion of the distribution function of the resonance excitations responsible for Landau attenuation. The nature of this distortion is such that the absorption of sound in the metal decreases, and if the sound is intense enough the absorption may become much smaller than the linear attenuation. This phenomenon, which was predicted theoretically in Ref. 1 and has been observed experimentally[2] in pure gallium, has come to be called momentum nonlinearity (MN). A necessary condition for the observation of MN is that the parameter $a=(\omega_0\tau)^{-1}$ be small, where τ is the excursion time of an excitation from its state of definite momentum and $\omega_0=q(\Phi/m)^{1/2}$ is the vibrational frequency of an electron trapped in the potential well produced by the wave (q is the wave vector of the sound, Φ is the amplitude of the sound-wave potential, and m is the effective mass of the electron). The condition $a \lesssim 1$ means that the electron succeeds in completing at least one oscillation before being scattered. According to the experimental data of Ref. 2, a rough estimate of the value of a that can be reached in ultrapure gallium specimens is ~0.5–1.0.

The large deviation of the electron distribution function from the equilibrium form produced by intense acoustic pumping may considerably alter the conditions for the propagation of a weak test signal whose velocity differs in magnitude and direction from that of the pump wave. Demikhovskiĭ and Maksimova[3] showed theoretically that both amplification and additional attenuation of the weak signal can be achieved in a pure metal under certain conditions. In the work reported here, this theoretical conclusion was investigated experimentally, and although "pure" amplification was not achieved, qualitative agreement of the observed phenomena with the theoretical predictions is demonstrated.

It has also been found experimentally[4,5] that, for a fixed wave intensity, the nonlinear decrease in the attenuation is considerably greater in the superconducting state than in the normal state. It has been hypothesized[5,6] that this effect may be associated with an anisotropic broadening of the superconducting energy gap for a group of resonance quasiparticles. Nevertheless, an unambiguous understanding of the reason for the existence of strong acoustic nonlinearity in superconductors has not yet been reached. In connection with certain new nonlinear phenomena predicted by Galaĭko and Shumeĭko[7] and specific for superconductors, we present in this paper some additional results that may help to clarify the experimental situation to some extent.

1. EXPERIMENTAL TECHNIQUE

All the experiments on nonlinear interaction of sound waves of various polarizations in a normal metal have been made using ultrapure gallium single crystals in which the electron mean free path l was ~1 cm; this appears to be the only metal in which MN has been well observed up to now. We also note that the nonlinear absorption due to dislocations was negligibly small in the investigated specimens.[1)] The conditions for the observation of nonlinear absorption in the superconducting state are not so rigid; in this case, not only have the above mentioned specimens been investigated, but also slightly doped (~0.01%) gallium, and aluminum with a residual resistance $R_{300}/R_{4.2}\sim 2\times 10^4$.

Figure 1 is a simplified block diagram of the experimental setup. The present experiment differs from previous ones[2,4] in that the test signal and pump signal were fed to the specimen by different transducers; this provided greater flexibility in studying the nonlinear interaction of waves of different polarizations. As a rule, the test signal consisted of a continuous sequence of pulses, while the pump signal was additionally amplitude modulated (modulation depth 100%) at a low frequency by a square-wave potential of the "meander" type. Then, by virtue of the strobing system and the system of synchronous amplification and detection at the modulation frequency, the receiver registered only the nonlinear addition to the absorption of the test signal that arose from the action of the pump wave. The passive band-pass filters made it possible almost entirely to avoid any direct action of the pump pulses on the receiving system.

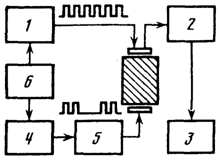

FIG. 1. Block diagram of the apparatus: 1—test signal generator, 2—band-pass filter at the test signal frequency, 3—receiver, 4—pumping signal generator, 5—band-pass filter at the pumping frequency, 6—pulsing unit.

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Since MN is sensitive to weak magnetic fields,[2,8] the measuring cell was enclosed by a double shield of permalloy and superconducting lead; this reduced the strength of the uncontrolled field at the specimen to 3–5 mOe. The magnetic field for the measurements was produced by a small single-layer superconducting solenoid mounted within the superconducting shield.

The specimens were immersed in liquid helium, whose temperature was measured with a carbon resistance thermometer located near the specimen. The signal from the thermometer was fed through a compensating system to one of the inputs of an $x-y$ plotter; the output voltage from the detecting system, calibrated with a $D4-3$ attenuator, was fed to the other input of the plotter.

A piezoelectric transducer consisting of a Y-cut lithium niobate crystal was used to generate both longitudinal and transverse sound waves in the specimen, the crystal being turned to the proper angle to assure mode operation.[9] The pulse length was ~1 μsec, and the highest pump intensities reached 30–40 W/cm^2 (the maximum powers were several times lower in the measurements on the superconducting state). The frequency range was 20–150 MHz.

2. EXPERIMENTAL RESULTS AND DISCUSSION

A. The normal state

We investigated the action of longitudinal and transverse pumping waves on the absorption of weak test waves for the cases in which the two interacting waves propagate in the same direction (parallel propagation) and in opposite directions (antiparallel propagation); test waves with both longitudinal and transverse polarization ε were used. With our experimental setup (Fig. 1) passage from the case of antiparallel propagation to that of parallel propagation was effected by time shifting the pumping wave with respect to the instant (taken as zero on the time scale) when the test signal entered the specimen. Thus, zero time corresponds to antiparallel propagation of the two signals, which encounter one another twice within the specimen. As the pump pulse delay is increased, the time during which the two waves interact while propagating in opposite directions decreases, and there is a smooth transition to the case of parallel propagation (when the two interacting waves have different polarizations, the difference between the propagation velocities of longitudinal and transverse oscillations must also be taken into account in interpreting the results).

The principal experimental facts observed in this study may be formulated as follows: 1) In antiparallel propagation, the attenuation of the test signal is always increased by the interaction between the two waves, regardless of their polarizations; 2) In parallel propagation with a longitudinal pumping wave and a transverse test signal, the attenuation of the latter is always reduced; the attenuation of a test signal having the same polarization as the pumping wave is also reduced; and 3) In parallel propagation of a transverse pumping wave and a longitudinal test signal, the nonlinear contribution to the attenuation of the latter may have either sign, depending on the ratio of the frequencies of the two waves, the pumping intensity, and the crystallographic orientation of the specimen. As a rule, the nonlinear addition is positive at the highest pumping intensity and frequency.

In Fig. 2, by way of illustration, we show typical plots of the nonlinear contribution to the attenuation of the test signal vs the pumping-wave delay time. The abscissae of the curves are normalized to the time for a single passage of the test signal through the specimen, and the ordinates are normalized to the linear absorption coefficient. The data for the transverse test signal were normalized only to the strain part of the linear attenuation coefficient since all effects associated with MN appear in a narrow phase region of momentum space near $\mathbf{q}\cdot\mathbf{v}_F=\omega$ ($\mathbf{v}_F$ is the Fermi velocity and ω is the frequency) with which only strain absorption is associated (the strain absorption was determined from superconductivity measurements). The characteristics of the specimens used to obtain the results discussed in this paper are listed in Table I.

Demikhovskiĭ and Maksimova[3] showed that in the case of substantial MN (when $a \ll 1$) the test-signal attenuation coefficient (only the strain part of the absorption was considered) propagating in a metal in the presence of a strong wave is given, to terms of the order of $\sim a$, by

$$\frac{\alpha}{\alpha_L}=\frac{4}{a\pi^2}\frac{S_1-S}{S}f\left(\frac{q_1}{q}\right)+O(a), \qquad (1)$$

where S_1 and q_1 are the velocity and wave vector of the weak wave, S and q are the corresponding quantities for the strong wave, and α_L is the linear attenuation of the weak wave. The analytic solution given in Ref. 3 was obtained only for integer and half-integer values of q_1/q; in this case $f(q_1/q)\sim 0.64$. It follows from Eq. (1) that with parallel propagation of two waves having different velocities one should observe amplification of the weak wave when $S_1<S$ and enhanced absorption of the weak wave when $S_1>S$. Under antiparallel propagation, the weak wave is always attenuated more

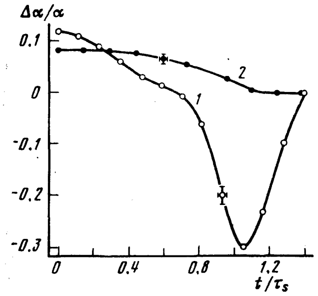

FIG. 2. Effects of waves with different polarizations in Ga on one another, q||a: 1—longitudinal pumping with a transverse test signal (f_{meas}=20 MHz, ε||c; f_{pump}=50 MHz, ε||a); 2—transverse pumping with a longitudinal test signal (f_{meas}=50 MHz, ε||a; f_{pump}=150 MHz, ε||b). The curves are normalized to the strain part of the absorption of the test signal and the times are expressed as multiples of the acoustic delay time of the test signal in the specimen.

TABLE I.

Direction $\mathbf{q}$	Polarization ε	α_Σ (cm·MHz)$^{-1}$ $\times 10^3$	a_D (cm·MHz)$^{-1}$ $\times 10^3$	$S \cdot 10^{-5}$, cm/sec	$\tau_S \cdot 10^6$, sec
$\mathbf{q} \parallel a$	$\varepsilon \parallel a$	10	10	4,322	2,7
$\mathbf{q} \parallel a$	$\varepsilon \parallel b$	22,7	4,4	2,810	4,2
$\mathbf{q} \parallel a$	$\varepsilon \parallel c$	4,79	3,17	2,747	4,3

Note: Here α_Σ is the total attenuation, α_D is the strain part of the attenuation, S is the velocity of sound, and τ_s is the time required for sound to pass once through the specimen.

strongly than linearly, regardless of the relation between S_1 and S. When $S_1 = S$ and the two waves propagate in the same direction, the first term on the right in (1) vanishes, and the remaining terms describe a decrease in the attenuation of the acoustic signal in the case of MN.[1]

Thus, if we speak only of the sign of the nonlinear contribution to the attenuation, we have qualitative agreement between the experimentally observed phenomena and the conclusions drawn in Ref. 3. But the uncertainty concerning the action of a transverse pumping wave on a longitudinal signal under parallel propagation is evidently associated with competition between the contributions from the two terms of opposite sign in (1), which differ in their a dependences. At the same time, we were unable to achieve "pure" amplification in the present work, so the question arises whether it is possible in principle to observe such an effect. Nevertheless, in our opinion there is some indirect confirmation of the existence of an "amplification" effect of the type described by Eq. (1). Figure 3 shows the magnetic-field dependence of the nonlinear addition to the absorption under the action of longitudinal pumping on a transverse signal for both parallel (curve 1) and antiparallel (curve 3) propagation, as well as the absorption of a strong transverse signal whose intensity is at least as high as that of the longitudinal pumping wave. It is well known[2,8] that a weak magnetic field almost entirely suppresses the MN, so the effect of a weak magnetic field on the absorption of a strong signal yields the magnitude of the strictly MN contribution, i.e. an estimate of the terms of order a in Eq. (1). It is evident from Fig. 3 that the nonlinear decrease in the attenuation of a transverse signal in longitudinal pumping (curve 1) is considerably greater in absolute magnitude than the action of transverse pumping on a transverse signal (curve 2). Similarly, Fig. 4 shows the action of longitudinal pumping on transverse (curve 1) and longitudinal (curve 2) test signals. In this case, too, longitudinal pumping acts considerably stronger on a transverse signal than on a longitudinal one; this provides qualitative confirmation of the validity of Eq. (1).

Formula (1) was derived under the assumption that $a \ll 1$, whereas only the weaker condition $a \lesssim 1$ could be satisfied in the experiments, so that a quantitative verification of the formula under these conditions would not seem to carry much weight. However, it may be assumed that when $S_1 \neq S$ the first term in (1) will give a functionally correct description of the principal nonlinear contribution to the attenuation of a test signal over a wider range of pumping powers, the more so that this term, as must be expected, vanishes in the limit $a \to \infty$. Since $\Phi = \Lambda_q U$ (Λ is the strain potential and U is the amplitude of the pumping wave), the nonlinear part of the absorption should be linear in $U^{1/2}$. Actually, this dependence is fairly well confirmed experimentally except for small values of U (curve 1 in Fig. 4). We also note that in the case of the action of a longitudinal pumping wave on a longitudinal test signal with parallel propagation [i.e., when the first term in Eq. (1) is absent] the amplitude dependence clearly differs from the square-root law (curve 2).

Since the quantity $f(q_1/q)$ in (1) is constant, $\Delta\alpha/\alpha_L$ should be independent of the frequency of the test signal provided the pumping conditions remain unchanged; this was verified experimentally for test signal frequencies of 20, 66, and 160 MHz at a fixed pumping frequency of 50 MHz under conditions in which an amplification type effect is observed (the requirement on the smallness of a is less rigid under these conditions than under conditions in which the attenuation is enhanced[3]). The maximum value of $\Delta\alpha/\alpha_L$ achieved was -0.3 ± 0.02; a quantitative estimate from the first term in (1) then gives $a \sim 0.6-0.7$, and this is in reasonable agree-

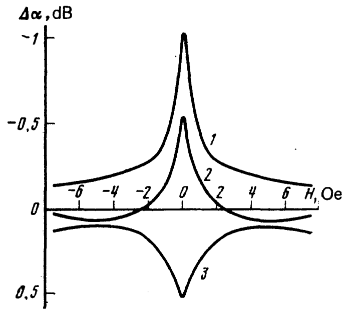

FIG. 3. Plots of the nonlinear contribution to the absorption of a test signal in Ga, $\mathbf{q} \parallel \mathbf{a}$, vs. the strength of an applied magnetic field ($f_{meas} = 66$ MHz, $\varepsilon \parallel \mathbf{c}$; $f_{pump} = 50$ MHz, $\varepsilon \parallel \mathbf{c}$): 1—parallel propagation of a transverse test signal and a longitudinal pumping wave, 2—self action of a strong transverse signal, 3—antiparallel propagation of a transverse test signal and a longitudinal pumping wave.

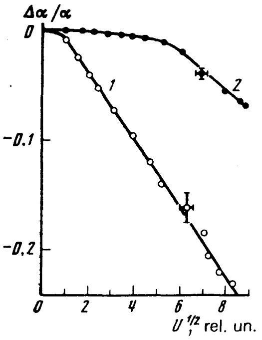

FIG. 4. Amplitude dependence of the nonlinear addition to the attenuation of a test signal under longitudinal pumping in Ga, $\mathbf{q} \parallel \mathbf{a}$ ($f_{pump} = 50$ MHz, $f_{meas} = 20$ MHz): 1—transverse test signal, $\varepsilon \parallel \mathbf{c}$, 2—longitudinal test signal, $\varepsilon \parallel \mathbf{a}$. The curves are normalized to the strain part of the test signal absorption.

ment with other estimates.

At least two reasons can be suggested for the inability to achieve "pure" amplification. First, the quantity a is not small enough; a rough estimate from Eq. (1) indicates that the condition $a \sim 0.1-0.2$ is necessary. The value of a can be decreased by further improving the purity of the metal, and also by substantially increasing the pumping frequency and power (these increases must be substantial because of their square-root contribution to a). Second, weak overlap in momentum space of the effective regions for the interaction of longitudinal and transverse waves propagating in symmetry directions, as they did in the present experiments, must also be considered as a possible reason. In this case, the strain absorption of transverse sound at central sections of the Fermi surface (FS) vanishes (because of the symmetry, the corresponding components of the strain potential tensor vanish), and all the attenuation is due only to the central sections of the FS, which are determined by the condition $\mathbf{q} \cdot \mathbf{v}_F = \omega$. To these parts there correspond a different rounded-off sections of the FS. At the same time, in the case of longitudinal oscillations the central sections of the FS, which also satisfy the conditions $\mathbf{q} \cdot \mathbf{v}_F = \omega$, may make the decisive contribution to the total attenuation. A more substantial overlap of the effective interaction regions could evidently be achieved by using an appropriate nonsymmetric direction, but a reasonable choice of such a direction would require detailed knowledge of the strain potential tensor over the entire FS.

B. The superconducting state

Nonlinear phenomena, including the decrease in the attenuation of a test signal under the action of pumping, become much more pronounced in the superconducting state. As was noted in a previous paper,[5] the nonlinear absorption observed in superconductors can hardly be associated with ordinary MN since it is observed in specimens in which the MN is completely suppressed by impurities (e.g., in aluminum).

In the work reported here we made a supplementary study of nonlinear effects in the superconducting state. By recording the purely nonlinear addition to the absorption we were able to study the nonlinear absorption of sound near T_c, as well as the amplitude dependence of the effect. Longitudinal oscillations were mainly used in all the measurements made in the superconducting state, since the superconducting transition curves for such oscillations have a simpler form (the region of rapid fall of the absorption characteristic of the attenuation of transverse sound is not present in the case of longitudinal sound).

Figure 5 shows recorded curves of the linear (curve 1) and nonlinear (curve 2) attenuation of the test signal in the presence of pumping of the same type under conditions of parallel propagation in pure gallium with $\omega\tau \sim 5-10$. We note three circumstances that in our opinion are important.

1) MN is present in the normal state.[2)] 2) Both the

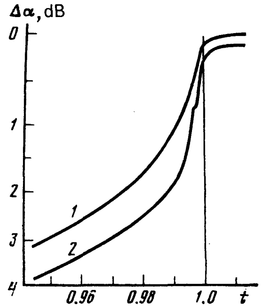

FIG. 5. Plots of the absorption coefficient of a test signal in superconducting Ga vs. temperature ($t = T/T_c$), $\mathbf{q} \| \mathbf{a}$ ($f_{meas} = 50$ MHz, $f_{pump} = 150$ MHz): 1—under linear conditions, 2—under nonlinear conditions.

linear- and nonlinear-attenuation curves have above T_c an exponential "tail"[10] whose origin is not entirely clear. Khlyustikov and Khaikin[11] observed similar behavior of the magnetic susceptibility in tin and attributed the presence of the "tail" to dislocations. Although the results of acoustic experiments do not agree in all respects with the dislocation idea, this evidently plays no important part in the present context: what is important is the existence of a temperature interval in which the degree of superconducting ordering is low. 3) The nonlinear-attenuation curve has near T_c a singularity that is reminiscent of a "step." Although this singularity is small, it is observed reliably enough at high pumping levels.

The behavior of all the observed features can be investigated more reliably by employing the method described above to record the strictly nonlinear contribution to the absorption. This is demonstrated in Fig. 6, where the zero point on each curve, i.e. the level at which the nonlinear contribution to the attenuation vanishes, is marked by an arrow. A weakly temperature dependent nonlinear contribution associated with MN becomes appreciable at temperatures far enough above T_c. At low pumping levels the absorption begins to fall even in the region of the "tail," and the "step" is not observed; this means that the effect of superconducting ordering on the attenuation of sound increases with pumping over the entire temperature range. As the pumping amplitude is increased the

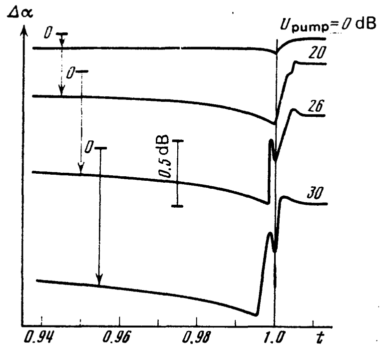

FIG. 6. Plots of the nonlinear addition to the attenuation of a test signal in Ga ($\mathbf{q} \| \mathbf{a}$) vs. temperature for several pumping amplitudes ($f_{meas} = 50$ MHz, $f_{pump} = 150$ MHz). The zero level is marked for each curve by an arrow.

"tail" begins to be suppressed (this corresponds to a decrease in the nonlinear contribution to the attenuation in relation to the MN level in the normal metal), and a peak corresponding to the "step" on Fig. 5 appears on the curve. Generally speaking, the deviation of the difference curves toward lower values of the nonlinear absorption at the superconducting transition might be attributed to a shift of the perturbed transition curve toward the lower temperatures as a result of overheating of the specimen with respect to the helium bath, since it was the temperature of the bath that was actually measured. However, the temperature at which the peak appears does not depend on the pumping power; this indicates that there is no appreciable overheating of the specimen and, consequently, that the depression of the "tail" is not associated with overheating, but is a nonlinear effect. Far from T_c the nonlinear addition to the absorption also varies weakly with temperature.

Figure 7 shows the dependence of the nonlinear contribution to the attenuation on the pumping amplitude for both the normal and the superconducting states, the curves being normalized to the linear attenuation at the given temperature. In a superconductor, this dependence is linear at low pumping levels but deviates from the linear trend at high pumping levels.

Figure 8 shows plots of the nonlinear addition to the absorption in aluminum. The values of $\omega\tau$ for the specimen is estimated to be no higher than 0.5. No MN is observed in the normal state, the positive nonlinear contribution to the attenuation being evidently associated with dislocations; in any case, it is not affected by a magnetic field. No phenomena were found in the superconducting state similar to the appearance of the "step" in the case of pure gallium. At the same time, the behavior of the curves far from T_c is very reminiscent, except for the relative magnitude of the effect, of the behavior of pure gallium. The absolute attenuation is greater than in gallium; this leads to slight overheating at high pumping levels, which manifests itself as a positive spike near T_c. The inset in Fig. 8 shows the amplitude dependences of the nonlinear contribution to the attenuation, normalized as in Fig. 7. The points cluster well about straight lines. Similar behavior has been observed in gallium specimens slightly doped

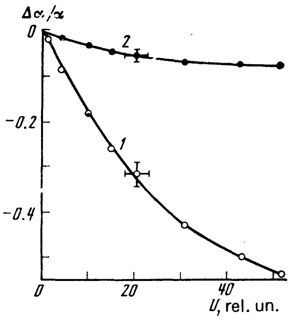

FIG. 7. Dependence of the nonlinear addition to the attenuation of a test signal on the pumping amplitude in Ga, q||a (f_{meas} = 50 MHz, f_{pump} = 150 MHz): 1—superconducting state ($\Delta\alpha_s/\alpha_s$, t = 0.94), 2—normal state ($\Delta\alpha_n/\alpha_n$, t = 1.1).

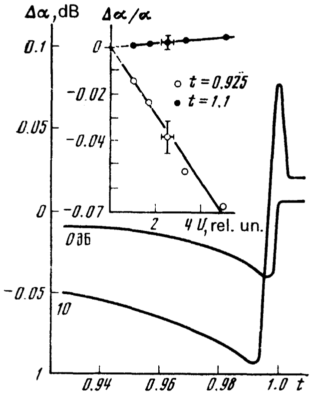

FIG. 8. Plots of the nonlinear absorption in Al vs. temperature (f_{meas} = 50 MHz, f_{pump} = 150 MHz). The inset shows the amplitude dependence of the effect: •—$\Delta\alpha_n/\alpha_n$, t = 1.1; ○—$\Delta\alpha_s/\alpha_s$, t = 0.925.

so as to reduce $\omega\tau$ to values below unity.

Galaĭko and Shumeĭko[7] called attention to a specific nonlinearity mechanism that should operate in a superconductor via the binding of excitations as a result of Andreev reflection of the excitations from the potential surface produced by a sound wave. Since the momentum of an excitation changes only slightly under Andreev reflection, the binding of quasiparticles by an acoustic field can take place at lower amplitudes than those required for MN. The amplitude and temperature dependences of these nonlinear phenomena are very complex. Under certain conditions nonlinearity of the "step" type should be observed near T_c, and very close to T_c, where $\Delta(T) \ll \Phi$ (Δ is the width of the energy gap) superconducting ordering should not affect the attenuation of sound at all. Thus, the features of nonlinear attenuation observed experimentally near T_c are in qualitative agreement with the conclusions drawn in Ref. 7.

The manifestation of the above noted nonlinearity far from T_c is reminiscent of ordinary MN with a threshold for the wave potential amplitude of $\Phi_0 \sim T_c/\omega\tau$. At large values of $\omega\tau$ this threshold can be considerably lower than the MN threshold, and one might attempt to attribute the observed increase in the nonlinear contribution to the attenuation in a pure superconductor to just this circumstance. Then one would have to suppose that under antiparallel propagation of the two signals one would observe an effect of the same sign as in the normal state but of greater magnitude. However, the experimental situation in this case is considerably different (Fig. 9): under antiparallel propagation of the test and pumping waves the effect changes sign immediately below T_c and the curves are very reminiscent of the case of parallel propagation. We also note that these curves exhibit no feature associated with the "step." In addition, it would seem quite unlikely that Andreev nonlinearity would appear in the complete absence of MN when $\omega\tau \lesssim 1$, since in this case the threshold amplitudes should be of the same order. Thus, an attempt to interpret the main nonlinear contribution to sound attenuation in a superconductor far from T_c from the point of view of Ref. 7 encounters certain difficulties, among which should be included the

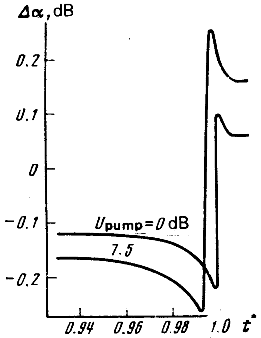

FIG. 9. Plots of the nonlinear attenuation of sound in Ga with antiparallel propagation of the test signal and pumping wave, q||a (f_{meas} = 50 MHz, f_{pump} = 150 MHz).

existence of an effect at small values of $\omega\tau$, and the fact that this effect has the same sign under parallel and antiparallel propagation of the test and pumping waves.

3. CONCLUSION

Let us formulate the principal results obtained in the present work.

The nonlinear interaction in a normal metal under MN conditions of waves having different polarizations is in qualitative agreement with the theoretical predictions of Ref. 3, although "pure" amplification was not achieved.

When $\omega\tau > 1$ nonlinear effects specific for superconductors appear near T_c, which consist in the suppression of the effect of weak superconducting ordering on acoustic attenuation and in the appearance of a "step" type feature on the curve. These phenomena agree qualitatively with the predictions of Ref. 7.

The nonlinear contribution to the absorption of a test signal in a metal in the superconducting state far from T_c is linear in the pumping amplitude provided the latter is not too high.

The results of the present work provide no grounds for rejecting the previously advanced[5,6] hypothesis of an anisotropic broadening of the energy gap of a superconductor under the action of acoustic pumping.

In concluding, we sincerely thank V.Ya. Demikhovskiĭ and V.M. Maksimova for a stimulating discussion before the experiments were formulated, and Yu.M. Gal'perin, B.I. Ivlev, V.I. Kozub, V.P. Semiozhenko, and V.S. Shumeĭko for many valuable discussions.

[1)]One can verify this by applying to the specimen a weak magnetic field that suppresses the NM completely[2] but does not alter the nonlinearity of dislocation origin.

[2)]One can verify this by applying a weak (~1 Oe) magnetic field, which suppresses the MN.

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