Absorption of longitudinal high frequency acoustic waves in $Y_{3-x}Lu_xAl_5O_{12}$ crystals

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The absorption of longitudinal high frequency acoustic waves in $Y_{3-x}Lu_xAl_5O_{12}$ ($0 \leqslant x \leqslant 3$) crystals is investigated theoretically and experimentally at temperatures $T < 80$ K in the case in which the absorption in a pure crystal is due to three-phonon processes. It is shown that the absorption of acoustic waves depends essentially on the impurity density. It is established that the frequency dependence of the sound absorption at low temperatures possesses a number of peculiarities. The form of the dependence corresponds qualitatively to that predicted theoretically.

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§1. INTRODUCTION

It is well known that the absorption of acoustic waves in dielectric crystals depends on the presence of impurities in them. Thus, for example, the lattice absorption of longitudinal waves at room temperatures in Si crystals alloyed with Ge is relatively insensitive to the presence of impurities; however, the absorption of transverse waves and the thermal conductivity of the crystal changes significantly in comparison with the corresponding values for an ideal crystal.[1,2]

The purpose of the present work is a theoretical and experimental investigation of the absorption of longitudinal high-frequency acoustic waves in $Y_{3-x}Lu_xAl_5O_{12}$ crystals at low temperatures, when the condition $\omega_s \tau_f > 1$ (ω_s is the frequency of the wave, τ_f is the phonon-phonon relaxation time).

In pure $Y_3Al_5O_{12}$ and $Lu_3Al_5O_{12}$ crystals (see Ref. 3), in the low temperature region ($\omega_s \tau_f > 1$) the frequency and temperature dependence of the absorption coefficient Γ of the longitudinal wave is described by the law $\Gamma \sim \omega_s T^n$($n \lesssim 4$, T is the temperature of the crystal), i.e., the absorption is determined fundamentally by three-phonon processes of the form $L + L \rightarrow L$.

In alloys of yttrium-aluminum garnet with rare-earth metals, the density and the elastic moduli of the crystal (at least, the elastic moduli of second order) change insignificantly.[4,5] The observed change in the character of the absorption (see below) can be connected with the significant change in the character of the kinetic processes in the crystals with impurities and, possibly, with the change in the phonon spectrum in comparison with the spectrum of the ideal crystal (see, for example, Ref. 6). At low concentrations of the impurity, $x \rightarrow 0$ and $x \rightarrow 3$, and for not too low temperatures ($\omega_s \tau_f \gg 1$) the indicated rearrangement of the phonon spectrum in a crystal with impurities can be neglected.

In the present work, we have limited ourselves to the study of absorption in $Y_{3-x}Lu_xAl_5O_{12}$ crystals in the temperature region $T < 80$ K, in which case the absorption is basically due to the interaction of the wave with acoustic phonons.

At higher temperatures it is necessary to take into consideration in the crystals studied the interaction of the waves with optical phonons, whose lower branches of which correspond[7] to energies ~ 170 K for $Y_3Al_5O_{12}$ and ~ 130 K for $Lu_3Al_5O_{12}$ (at the center of the Brillouin zone). Beginning with the indicated energies, the phonon density of states of the crystal increases sharply,[5] and if these states are occupied the absorption can be determined, to a significant degree, by interaction with the optical phonons.

§2. THEORY

The effect of impurities on the kinetic processes in a dielectric crystal, which determine the absorption, has been investigated in a number of researches.[6–13] The case of low frequency waves was considered in Refs. 8–12. A more general approach, which is applicable to the absorption of high-frequency waves, was developed in Ref. 13, where it is assumed that the effect of the impurities on the kinetic processes in a system of thermal phonons can be described by use of the quasielastic phonon-impurity relaxation time (in the general case, with the aid of some sequence of relaxation times τ_n).

The expression for the absorption coefficient has the form

$$\Gamma = \frac{C_v T \omega_s}{2\rho v_s(qJ)} \operatorname{Re}\Big\{ i\langle \gamma_s^2(kj) P(kj) \rangle - \frac{1}{\omega_s} \cdot \frac{\langle \gamma_s(kj) \tau_f^{-1}(kj) P(kj) \rangle^2}{\langle \tau_f^{-1}(kj) [1 - i\omega_s^{-1} \tau_f^{-1}(kj) P(kj)] \rangle} \Big\} \qquad (1)$$

where

$$P(kj) = \frac{i\omega_s \tau_0(kj)}{1 - i\omega_s \tau_0(kj) \alpha_{Jj} B(kj)},$$

$$B = -w_{f0} - \cfrac{1^2}{-3w_{f1} - \cfrac{2^2}{-5w_{f2} - \cfrac{3^2}{- \dots}}},$$

$$w_{fn} = \left(1 + \frac{i}{\omega_s \tau_{fn}}\right) \frac{1}{\alpha_{Jj}}, \quad \tau_{fn}^{-1} = \tau_f^{-1} + \tau_n^{-1}, \quad \alpha_{Jj} = \frac{v_s(j)}{v_s(J)}.$$

Here τ_n is the sequence of relaxation times of the harmonics of the perturbed phonon distribution function in the case of the scattering of the phonons by impurities, v_s is the velocity of the wave, J and j denote the polarizations of the acoustic and thermal phonons, C_v and ρ are the specific heat and the density of the crystal, $\gamma_s(kj)$ is a symmetrized combination of the Grüneisen constants for phonons of the kjth mode, $\langle \ldots \rangle$ is the symbol of kinetic averaging by the standard procedure.

The expression (1) has a rather general character. In particular, for a pure crystal, when $\tau_0 \rightarrow \infty$, (1) gives the correct results both for the absorption of low frequency ($\omega_s \tau_f \ll 1$) and for the absorption of high-frequency ($\omega_s \tau_f \gg 1$) waves. The results of Refs. 8–12 follow from (1) with account of the corresponding assumptions. For a number of limiting cases of absorption of a longitudinal wave propagating in a dielectric crystal with a large number of impurities ($\tau_0 < \tau_f$), explicit relations can be obtained:

a) in the region of extremely low frequencies, at $\omega_s < \tau_f^{-1}$, we have $\Gamma \propto \omega_s^2 \tau_f$;

b) at $\tau_f^{-1} < \omega_s < (\tau_f \tau_0)^{-1/2}$ we have $\Gamma \propto \tau_f^{-1}$;

c) at $(\tau_f \tau_0)^{-1/2} < \omega_s < \tau_0^{-1}$ we have $\Gamma \propto \omega_s^2 \tau_0$;

d) in the region of extremely high frequencies, at $\omega_s > \tau_0^{-1}$, we have $\Gamma \propto \omega_s$.

The following explanation can be given for these results. The absorption coefficient is determined by the ratio of the rate of entropy production in a phonon system to the flow of energy in the wave. In the propagation of a wave in a crystal, the nonequilibrium character of the phonon distribution function is determined by modulation of the eigenfrequencies of the phonons of the wave and modulation of the temperature of the phonons. For relaxation of the phonons to a state described by some local temperature, quasielastic processes are sufficient, while the relaxation of the phonons to the equilibrium (with temperature of the thermostat) distribution is determined by inelastic processes. In crystals with a large number of impurities ($\tau_0 \ll \tau_f$) quasielastic relaxation takes place rapidly. The contribution of the quasielastic processes to the rate of entropy production is additive and can always be separated. Over the entire frequency range $\omega_s < \tau_0^{-1}$ the absorption coefficient contains a term proportional to $\omega_s^2 \tau_0$ and governed by quasielastic processes. This term is decisive if the modulation of the temperature of the phonons by the acoustic wave is insignificant, as, for example, in the range $(\tau_f \tau_0)^{-1/2} < \omega_s < \tau_0^{-1}$, where the length of the absorbed wave becomes small in comparison with the characteristic length of relaxation of the energy of the phonons in the impure crystal $l \sim v_s (\tau_0 \tau_f)^{1/2}$. In the region of lower frequencies, the modulation of the temperature of the phonons of the longitudinal wave becomes significant and in the expression for the rate of entropy production a contribution that depends on τ_f appears from the inelastic processes. For extremely low frequencies $\omega_s < \tau_f^{-1}$ this contribution is $\tau_f / \tau_0 \gg 1$ times the contribution from the quasi-elastic relaxation; therefore the absorption coefficient $\Gamma \propto \omega_s^2 \tau_f$.

The most interesting is the region of frequencies $\tau_f^{-1} < \omega_s < (\tau_f \tau_0)^{-1/2}$. At $\omega_s > \tau_f^{-1}$ in an ideal crystal, the absorption is determined by the group of resonant phonons belonging to a certain surface in the space of wave vectors, singled out by the selection rules in the case of phonon-phonon collisions. In crystals with a large number of impurities, the quasielastic collisions of the phonons with the impurities are frequent in the considered range of frequencies, and it is not possible to consider the absorption in terms of three-phonon collisions. As before, the distribution of the phonons is locally connected with the deformation in the wave; however, the rate of entropy production in the system changes as a consequence of the ineffectiveness of the phonon-phonon collisions because of the temporal dispersion. As before, the relaxation formula for the absorption coefficient

$$\Gamma \propto \omega_s^2 \tau_f / (1 + \omega_s^2 \tau_f^2)$$

remains valid and Γ is determined by the time of the inelastic processes τ_f. The absorption coefficient does not depend on the frequency and is characterized by a strong temperature dependence $\Gamma \propto C_v T \tau_f^{-1}(T)$.

The presence of two relaxation times of the acoustic phonons leads to the result that the absorption in the frequency range $\tau_f^{-1} < \omega_s < \tau_0^{-1}$ turns out to be smaller than in the ideal crystal ($\tau_f < \tau_0$), and this decrease in the absorption is not connected with the change in the elastic properties of the material by the alloying, properties not taken into account in Ref. 13.

In the frequency range $(\tau_f \tau_0)^{-1/2} < \omega_s < \tau_0^{-1}$ the decrease in Γ in the alloyed crystal, in comparison with that in the unalloyed, is greater the smaller the ratio τ_0 / τ_f. If τ_f changes weakly with increase in the impurity density, the indicated ratio has a minimum for a definite composition. This means that at a fixed frequency of the wave in the range $(\tau_f \tau_0)^{-1/2} < \omega_s < \tau_0^{-1}$ the curve of the dependence of Γ on the concentration of substitution impurities has a minimum.

§3. EXPERIMENT

The effect of impurities on the lattice absorption of the acoustic waves has been studied experimentally in a system of solid solutions of yttrium- (lutecium) aluminum garnets (YAG–LAG). Single crystal samples of $Y_{3-x}Lu_xAl_5O_{12}$ ($0 \leqslant x \leqslant 3$) were prepared by the method of horizontal directed crystallization in a molybdenum container. The closeness of the values of the ionic radii of yttrium and lutecium contributed to their mutual isomorphic substitution, with a distribution coefficient very close to unity. The concentrations of Y and Lu in the garnet were estimated from the density and from x-ray measurements of the lattice constant.[14]

For acoustic measurements, samples of size $0.4 \times 0.4 \times 1.0$ cm were cut from the ingots. They were oriented along the [110] axis. For excitation and detection of longitudinal waves, grain-oriented piezoelectric films of zinc oxide were deposited on the plane-parallel polished ends of the sample by the method of magnetron sputtering. The broad-bandedness of the resulting transducers made it possible to carry out acoustical measurements, with a single sputtering of the transducer, over the range from 0.5 to 9.4 GHz.

It is first appropriate to show that in the system of solid solutions studied, because of the presence of Lu impurities in YAG and Y impurities in LAG, a change takes place in the relaxation time τ of the thermal phonons. Figure 1 shows the frequency dependences of the absorption at liquid nitrogen temperature; the curves have a sharply pronounced transi-

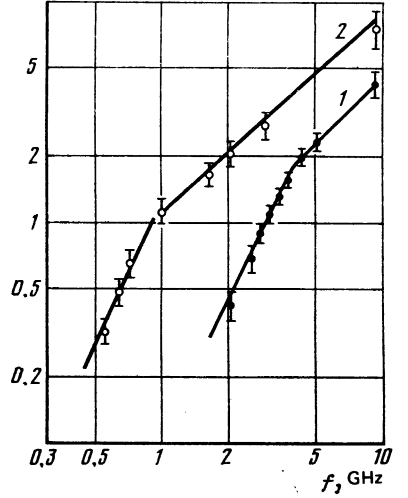

FIG. 1. Frequency dependence of absorption at $T = 77$ K for two compositions of a solid solution: curve 1—$Y_{2.7}Lu_{0.3}Al_5O_{12}$; curve 2—$Y_1Lu_2Al_5O_{12}$.

tion from a quadratic to a linear dependence on the frequency. The bend for the case of ideal crystals was connected in Refs. 15 and 16 with the transition from the Akhiezer absorption mechanism to absorption in three-phonon processes at $\omega_s\tau \sim 1$. In the case of impure crystals, the approach of the absorption to a dependence $\propto \omega_s^{-1}$ undoubtedly corresponds to the absorption mechanism in three-phonon processes. However, the transition to the quadratic frequency dependence of the absorption and the associated inflection point $\omega_s\tau \sim 1$ can correspond in the general case, in the framework of the model of Ref. 13, to the time of quasielastic scattering of the thermal phonons by impurities in the crystal. The dependence of the time τ thus determined on the composition of the solid solution is shown in Fig. 2. The curve in Fig. 2 allows us to state unequivocally that the scattering centers, i.e., impurities affect the relaxation of the thermal phonons. Moreover, the approximate equality of the value of τ at $x = 0$ and $x = 3$, when the relaxation of the phonons is controlled by the phonon-phonon interaction, i.e., by τ_f, means that in the case of a complete isomorphic substitution Y⇄Lu in the lattice of the garnets, significant change in the character of the phonon-phonon interaction are not observed, as was to be expected.

The effect of scattering of the photons by the impurities manifests itself in our experiments in a decrease in the absorption at room[16] and liquid-nitrogen temperatures. Nevertheless, the maximum effect is observed at $T \approx 30$–40 K,[18] when the difference between τ_0 and τ_f is particularly strong.

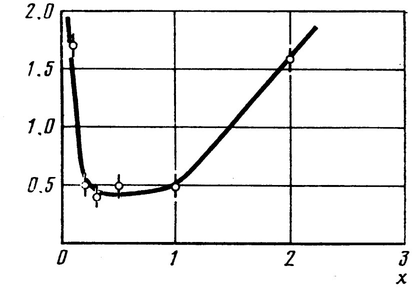

FIG. 2. Dependence of the relaxation time of thermal phonons in $Y_{3-x}Lu_xAl_5O_{12}$ crystals ($0 \leqslant x \leqslant 3$) at $T = 77$ K.

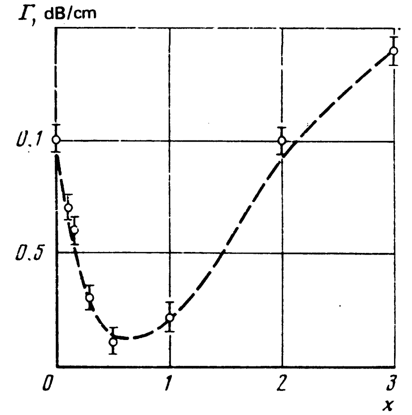

FIG. 3. Dependence of the absorption on the composition of the solid solution ($T = 40$ K, $f = 2.5$ GHz).

Typical results of the dependence of the absorption difference $\Gamma - \Gamma_{4.2\ \mathrm{K}}$ on the composition of the solid solution measured at a frequency of 2.5 GHz and at $T = 40$ K are shown in Fig. 3. $\Gamma_{4.2\ \mathrm{K}}$ is the value of temperature-independent residual losses due to structural and structural crystal imperfections,[19] usually subtracted out in the study of absorption in dielectrics. The curve in Fig. 3 has a broad minimum, very similar to the curve of the dependence of τ on the composition of the solid solution (see Fig. 2). Such a form of the dependence of Γ at temperatures that are charcteristic for the absorption mechanism in three-phonon processes $\omega_s\tau_f \gg 1$ shows $\Gamma \propto \tau$ in the case of impure crystals, even at

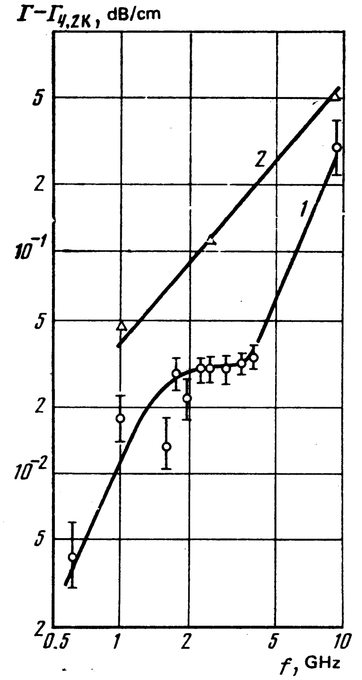

FIG. 4. Dependence of the absorption on the frequency at $T = 40$ K: curve 1—$Y_{2.7}Lu_{0.3}Al_5O_{12}$, curve 2—$Y_3Al_5O_{12}$.

rather low temperatures, which agrees with the results of Ref. 13.

The frequency dependence of the absorption Γ at $T = 40$ K turned out to be especially interesting. It is shown in Fig. 4. Curve 1 in Fig. 4 refers to a crystal of composition $Y_{2.7}Lu_{0.3}Al_5O_{12}$, for which the quantity τ from Fig. 2 is close to minimal. For comparison, the absorption curve for pure YAG crystal is shown also in this graph. Over the entire range of frequencies studied, there is not only an overall decrease in the absorption but also a change in its character. Thus, in the frequency range 1–3 GHz, a flattening of the plot is seen, while to the right and left of this anomaly the dependence corresponds to the law $\Gamma \propto \omega_s^n$, where $n \lesssim 2$. A similar character of $\Gamma = f(\omega_s)$ follows from Ref. 13.

§4. DISCUSSION OF RESULTS

1. The experimental results indicate that the presence of impurities in the crystal has a significant effect on the value and character of the absorption of longitudinal waves, especially under low temperature conditions, when, for pure crystals, $\omega_s \tau_f \gg 1$, and the absorption is determined by three-phonon processes.

2. The dependence of the absorption coefficient of high-frequency sound on the composition of the solid solution has a minimum. This dependence is qualitatively identical to the similar dependences of the relaxation times of the phonon system, which are determined from the absorption (see Fig. 2) and from the thermal conductivity.[20] Thus, the presence of impurities significantly changes the kinetic processes in the phonon system, and in the frequency range studied the absorption is determined by some set of phonon relaxation times.

3. The frequency dependence of the absorption coefficient in $Y_{3-x}Lu_xAl_5O_{12}$ crystals at low temperatures is characterized by a number of features, the chief of which is its flattening out in a finite range of frequencies. The character of the frequency dependence agrees qualitatively with the character of the corresponding theoretical curve. We shall now discuss in detail the behavior of the inflection point in the case of a change in temperature of the crystal. The insignificant shift of the left inflection point in $Y_{2.7}Lu_{0.3}Al_5O_{12}$ at 40 K relative to the inflection point at 77 K (one might expect $\tau_f(T) \propto T^{-5}$ and the corresponding shift to be expressed more strongly) can be connected with the following: a) with the significant decrease in the inelastic relaxation time of the phonons in a crystal with a large number of impurities in comparison with the pure crystal, or b) with the impossibility of correctly taking into account the role of the optical phonons in sound absorption at high temperatures, in particular, at 77 K. The observed shift of the right-hand inflection point indicates an increase in the frequency of quasielastic scattering of the phonons. If the phonon scattering were by impurities, in the given case, by Lu atoms, it would possess a Rayleigh character; then the indicated inflection point would be displaced in the opposite direction, since in Rayleigh scattering, $\tau_0(T) \propto T^{-4}$. The shift of the right-hand inflection point toward higher frequencies upon decrease in the temperature is possibly connected with the formation of resonant vibration in the case of replacement of Y atoms by the heavy Lu atoms.

4. It has been shown experimentally that, upon introduction of rare-earth metals into the YAG lattice, the second-order elastic moduli remain virtually unchanged (within the limits of 2–3%).[4,5] There is no basis for assuming that the third-order elastic moduli change significantly. Moreover, a change in the third-order elastic moduli could only lead to a shift of the inflection point on the frequency dependence of the absorption coefficient, which would have the same form as for the ideal crystal, but not to the observed qualitative change of this dependence. Thus, at least for $x \to 0$ and $x \to 3$, the observed features of the absorption are connected with significant rearrangement of the kinetic processes in the phonon system in the presence of fast phonon relaxation processes in the quasimomentum, and to slow one in the energy.

5. For intermediate values of doping, the adequacy of the model of Ref. 13 of the experimental situation becomes unclear. This is due to the rearrangement of the phonon spectrum because of the formation of impurity vibrational states. The problem of absorption under these conditions therefore requires a special investigation.

[1] K. R. Keller, J. Appl. Phys. **38**, 3777 (1967).

[2] V. V. Lemanov, V. Ya. Avdonin and A. V. Petrov, Pis'ma Zh. Eksp. Teor. Fiz. **12**, 515 (1970) [JETP Lett. **12**, 360 (1970)].

[3] M. J. Dutoit, J. Appl. Phys. **45**, 28 (1974).

[4] V. F. Kitaeva, N. N. Sobolev, I. L. Chistyĭ, E. V. Sharikov, V. V. Osiko, M. I. Timoshechkin and A. S. Zolotko, Fiz. Tverd. Tela **22**, 1379 (1980) [Sov. Phys. Solid State **22**, 805 (1980)].

[5] S. I. Morozov, S. A. Danilkin, V. V. Zakurkin, S. N. Ivanov, V. V. Medved', S. F. Akhmetov and A. G. Davydenko, Fiz. Tverd. Tela **24**, 2568 (1982) [sic!].

[6] A. M. Kosevich, Fizicheskaya mekhanika real'nykh kristallov (Physical Mechanics of Real Crystals) Kiev, Naukova dumka, 1981.

[7] G. Mace, G. Schaack, N. G. Taaning and J. A. Konigstein, Z. f. Phys. **230**, 391 (1970).

[8] P. B. Miller, Phys. Rev. **137**, 1937 (1965).

[9] L. E. Gurevich and B. I. Shklovskiĭ, Zh. Eksp. Teor. Fiz. **53**, 1726 (1967) [Sov. Phys. JETP **26**, 989 (1968)].

[10] V. D. Kagan, Fiz. Tverd. Tela **23**, 1551 (1981) [Sov. Phys. Solid State **23**, 909 (1981)].

[11] H. J. Maris, Phys. Rev. **175**, 1077 (1968).

[12] Yu. A. Logachev, Fiz. Tverd. Tela **15**, 3454; **16**, 3109 (1973) [Sov. Phys. Solid State **15**, 2308; **16**, 2008 (1973)].

[13] Yu. V. Gulyaev, and A. G. Kozorezov, Zh. Eksp. Teor. Fiz. **82**, 1551 (1982) [Sov. Phys. JETP **55**, 898 (1982)].

[14] S. F. Akhmetov, G. L. Akhmetova, G. A. Gazizova and T. F. Mirenkova, Zh. Neorg. Khim. **22**, 2966 (1977).

[15] Yu. V. Ilisavskiĭ and D. Chiplis, Fiz. Tverd. Tela **14**, 2412 (1972) [Sov. Phys. Solid State **14**, 2085 (1972)].

[16] S. N. Ivanov, G. D. Mansfel'd and E. N. Khazanov, Fiz. Tverd. Tela **15**, 317 (1973) [Sov. Phys. Solid State **15**, 235 (1973)].

[17] S. F. Akhmetov, G. A. Gazizova, S. N. Ivanov, I. M. Kotelyanskiĭ, A. N. Makletsov, V. V. Medved' and V. N. Posadskiĭ, Fiz. Tverd. Tela **19**, 308 (1977) [Sov. Phys. Solid State **19**, 177 (1977)].

[18] A. P. Andreev, S. F. Akhmetov, A. G. Davydchenko, C. N. Ivanov, V. V. Medved', S. V. Kolodieva and I. M. Kotelyanskiĭ, *ibid.* **24**, 1228 (1982) [**24**, 696 (1982)].

[19] S. N. Ivanova and E. N. Khazanov, Radiotech. Elektron. **26**, 402 (1981).

[20] L. N. Vasil'ev, I. Dzhabbarov, V. S. Oskotskiĭ, L. S. Parfen'eva and I. A. Smirnov, Teploprovodnost' materialov, soderzhashchikh RZM. Tesisy VII Vsesoyouznoĭ konferentsii po teplofizicheskim svoĭstvam vesnchestv. (Thermal Conduction of Materials Containing Rare Earth Metals; Papers at the VII All-union Conference on thermophysical Properties of Materials), Tashkent, 1982.

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