revista mexicana de física 26 no. 2 (1979) 233-254

PHONONS AND SUPERCONDUCTIVITY: RECENT DEVELOPMENTS

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ABSTRACT

In this article we review recent developments which relate the spectral electron-phonon density $\alpha^2(\omega)F(\omega)$ - fundamental to the Eliashberg formulation of superconductivity - to the inverse lifetime of phonons $\Upsilon_{\lambda}(q)$. Neutron scattering experiments give not only the energy of the phonon $h\omega_1(q)$ (center of the group) but also the width of the group $\Gamma_{\lambda}(q)$. For Nb, measurements of $\gamma_{\lambda}(q)$ and calculations of phonon lifetimes which take into account the band structure have produced new information on $\alpha^2(\omega)F(\omega)$. After a brief review of the tunneling technique which leads directly to $\alpha^2(\omega)F(\omega)$ we turn to the discussion of the problem of soft phonons in compounds and their possible relationship to high T_c . Exploiting the available information on $\alpha^2(\omega)F(\omega)$ for Nb₃Sn we give a quantitative argument which demonstrates that low frequency modes are unimportant in this case. We also present qualitative arguments confirming the predictions of calculations of the functional derivatives which show that the most effective energies for high $T_{\rm c}$ are intermediate ones, of the order of 7KBTc.

In the final section we describe recent work on phonon widths in the superconducting state. The electrons condense in Cooper pairs and therefore if the phonon energy available is less than twice the gap these particular phonons will not decay through electron-phonon interaction. We argue that this experiment could be a good probe of anisotropy in the superconducting state.

RESUMEN

En este artículo revisamos trabajos recientes que relacionan la densidad espectral electrón-fonón $\alpha^2(\omega)F(\omega)$ - fundamental a la formulación de Eliashberg de la superconductividad - con el inverso del tiempo de vida de los fonones $\gamma_{\lambda}(\underline{q})$. Los experimentos de dispersión de neutrones permiten medir la energía del fonón $\hbar\omega_{\lambda}(\underline{q})$ (centro del grupo) y, además, el ancho del grupo $\Gamma_{\lambda}(\underline{q})$. En el caso del Nb, medida de $\gamma_{\lambda}(\underline{q})$ y cálculo deltiempo de vida de los fonones, tomando en cuenta la estructura de bandas, han producido nueva información acerca de $\alpha^2(\omega)F(\omega)$.

Después de una revisión breve de la técnica de tunelamiento que permite conocer $\alpha^2(\omega)F(\omega)$ directamente, nos ocupamos del problema de los fonones blandos en compuestos y su posible relación con altas temperaturas críticas. Haciendo uso de la información conocida acerca de

 $\alpha^2 \left(\omega \right) F \left(\omega \right)$ para Nb₃Sn damos argumentos cuantitativos que demuestran que los fonones de baja frecuencia no son importantes en este caso. También presentamos argumentos cualitativos, confirmando las predicciones de los cálculos de las derivadas funcionales que muestran que las energías más efectivas para elevar la temperatura crítica son las intermedias, de orden $7 K_{\rm B} T_{\rm c}$.

En la sección final, describirmos trabajos recientes en el ancho de banda de los fonones en el estado superconductor. Los electrones se condensan en Pares de Cooper y, por lo tanto, si la energía disponible es menor que el doble del ancho de la banda superconductora, el fonón referido no puede decaer por interacción electrón-fonón. Consideramos que estos experimentos podrían servir para medir la anisotropía en el estado superconductor.

Simple s-p metals are made up, to a good approximation, of closed shell ion cores of charge +Z plus Z free electrons per ion which can easily move throughout the entire crystal lattice. The ion cores form a regular array (Fig. 1) with small excursions off



Fig. 1. Ion cores in a metal

equilibrium possible at finite temperature. For small vibrations of the ions the harmonic approximation is valid and the dynamics of the system of ions reduces to that of a system of 3N coupled harmonic oscillators. Here 3N is the number of independent degrees of freedom. The 3 gives the three directions (x,y,z) of motion and N is the number of ions.

The frequencies of the normal modes of vibration of the crystal lattice i.e. of the phonons can be measured quite accurately by the method of inelastic neutron scattering.

Cold neutrons available around high flux nuclear reactors are in a sense ideal probes of the phonons. The neutron is coupled directly to the nucleus at the center of each ion because of the nuclear force. If it is made to pass through a metal it can be inelastically scattered setting the ions in motion i.e. creating a normal mode of vibration. In the constant Q mode of operation of the triple axis spectrometer an incident beam of neutrons of known energy and momentum is scattered and only those neutrons having lost a definite momentum <u>k</u> (which is transferred to the ions) are selected from the scattered beam. These scattered neutrons are then analyzed in energy. The intensity I as a function of energy transfer E froms what is called a neutron group. As shown in Fig. 2 I(E) vs E is small everywhere except for a peak around some energy ω which is identified as the energy of the phonon of momentum <u>k</u> with the width of the group related to the phonon lifetime. (In practice multiple phonon processes complicate the picture).



Fig. 2. A neutron group

Thus we can obtain the dispersion curves for the phonons, i.e. ω vs <u>k</u> as shown schematically in Fig. 3. For each direction of k in a simple



Fig. 3. Dispersion curves for the phonons

metal like Pb there are three branches to the dispersion curves: ω_{λ} with $\lambda = 1, 2, 3$; one longitudinal and two transverse. Also the phonons range in energy from zero to 10 meV (10^{-2} eV) for Pb and <u>k</u> ranges over the first B.Z. only. This is a sufficient set of momentum labels to enumerate the 3N normal modes of vibration.

The reason that cold a neutron of energy \circ (a few meV) has a wavelength of the order of an interatomic spacing $(\circ A)$. Therefore, on creation of a phonon , the neutron loses an amount of energy and momentum of the order of the amount it had in the first place. This is clearly ideal for accurate measurements.

From the dispersion curves (one for each direction in the first Brillouin zone) we can construct the phonon frequency distribution $F(\omega)$ defined by

$$F(\omega) = \frac{1}{N} \sum_{\substack{\underline{k}\lambda \\ F \cdot B \cdot Z}} \delta(\omega - \omega_{\lambda}(\underline{k})).$$
(1)



Fig. 4. Phonon frequency distribution $F(\omega)$ as given by Cowley⁽⁹⁾

In (1) each frequency $\omega_{\lambda}(\underline{k})$ is, in turn, put into the Dirac δ -function which checks whether or not this particular ω_{λ} is equal to ω . This gives a histogram which tells us how many phonons there are between ω and $\omega + d\omega$. It must then be appropriately normalized. Results for Pb are given in Fig. 4.

While, to know the phonons in complete detail, it is necessary to go back to the dispersion curves (one picture per direction of <u>k</u> in the F.B.Z.) for some applications $F(\omega)$ (a single curve) is sufficient. For example, the thermodynamics of a phonon gas follows from $F(\omega)$ without reference to the dispersion curves. As a single more concrete example, the internal energy U is given by

$$U = \begin{cases} F(\omega)n(\omega)\omega d\omega \end{cases}$$
(2)

where $n(\omega)$ is the Bose distribution function at temperature T. Thus we can say that all of the complex information on the phonons contained in the dispersion curves can be condensed in to a single function $F(\omega)$ as far as themodynamics is concerned. This is a great simplification.

We now turn to the free electrons. Their most important property is that they are nearly free. They can easily be made to drift by a small external electric field \underline{E} leading to a macroscopic current

 $J = \sigma E \qquad (\sigma \text{-conductivity}) \tag{3}$

The ground state of a set of ZN free electrons is a Slater determinant of those plane waves $e^{i\underline{k}\cdot\underline{x}}$ with energy closest to zero with 2 electrons per state in accord with the Pauli exclusion principle. In momentum space (<u>k</u>) this defines a sphere of radius k_F (the Fermi momentum) of constant energy E_F (the Fermi energy) with all states inside occupied and all states outside unoccupied (Fig. 5). In a real crystal the Fermi surface is more complicated and the plane waves are to be replaced by Bloch states. These are details that don't bear directly on our present discussion.





The Fermi sphere describes well, at least in a first approximation, the normal state. When at low temperature Pb becomes a superconductor the free electrons undergo a phase transition to a new "super state". The electronic wave function of lowest energy is not the Fermi sphere but rather a superposition of Cooper pair wave functions. Each electron is now condensed into a "macromolecule" or condensate and a finite energy \triangle (the energy gap) is needed to break its hinding to the condensate. The exact nature of the wave function is described by B.C.S. theory but is too complicated to repeat here. It will not be necessary. What is important to know is that the condensation occurs if the net interaction between two electrons is attractive. This attraction comes from the electron-ion interaction i.e. the electron phonon interaction. As shown in Fig. 6 and electron can pull in the + ions around it increasing the ion density in that region. This increased density of + charge, in turn, attracts a second electron. Thus the pola-





rizable system of + ions provides a mechanism where by the Cculomb repulsion μ^* between two electrons might be reduced and, in fact, in some sense, overcome, leading to an effective attraction between two electrons. The matrix element of the electron-phonon interaction scattering an electron from k to k' due to the emission or absorption of a phonon $\omega_\lambda^{}\,(\underline{k}\,{}^t\,{}^-\underline{k})$ is denoted by $g_{k\,{}^t\,k\lambda}^{}$ and shown schematically in Fig. 7. The most important electrons in superconductivity (i.e. those most affected by the phase transition) are those at the Fermi energy $E_F = a$ few e.v. They scatter to a final state of energy -- $E_{\rm E} \pm \hbar \omega_{\chi}$ which does not differ in energy very much from $E_{\rm E}$. Thus we will be interested only in Fermi surface to Fermi surface transitions coming from the electron-phonon interaction $g_{k'k\lambda}$. In fact all of the complicated information on the electrons, the phonons and the electronphonon interaction can be condensed with no approximation into a single weighted phonon frequency distribution $\alpha^2(\omega)F(\omega)$ which describes completely the electron-electron interaction mediated by the phonon as it enters superconductivity. Thus function $\alpha^2(\omega)F(\omega)$ is

 $\alpha^{2}(\omega)F(\omega) = \frac{\sum_{\underline{k}\underline{k}'} |g_{\underline{k}'\underline{k}\lambda}|^{2} \delta(\varepsilon_{\underline{k}}) \delta(\varepsilon_{\underline{k}'}) \delta(\omega - \omega_{\lambda}(\underline{k}' - \underline{k}))}{\sum_{\underline{k}} \delta(\varepsilon_{\underline{k}})}$ (4) $\underbrace{\underline{k}'e^{-}}{\omega_{\lambda}(\underline{k}' - \underline{k})}$

Fig. 7. An electron scatters from $\underline{\kappa}$ to $\underline{\kappa}'$ due to the emission or the absorption of a phonon $\omega_{\lambda}(\underline{\kappa}' - \underline{\kappa})$

in which we repeat again, only Fermi surface to Fermi surface transitions are allowed and each phonon mode is weighted by the strength of $|g_{k'k\lambda}|^2$ for that transition.

In the Eliashberg formulation of superconductivity which is exact to order square root of electron m to ion M mass, i.e. $\sqrt{\frac{m}{M}}$, the critical temperature T_c (and all other properties of the superconducting state) is a functional of $\alpha^2(\omega)F(\omega)$ and μ^* . That is

$$T_{C} = F(\alpha^{2}(\omega)F(\omega), \mu^{*})$$
(5)

where the complicated nonlinear coupled integral equation of Eliashberg determines numerically the functional F. The details of these equations are not important to us. What is more important is that all the properties of the superconducting state follow from a knowledge of $\alpha^2(\omega)F(\omega)$ and μ^* .

Calculation of $\alpha^2(\omega)F(\omega)$ based on equation (4) were done as early as 1968 by Carbotte and Dynes⁽¹⁾ and more recently by Tomlinson and Carbotte⁽²⁾. These latest calculations use the real phonons revealed from inelastic neutron scattering and a 4-plane wave pseudopotential theory for the electronic wave functions. Results for $\alpha^2(\omega)F(\omega)$ in the case of Pb look very much the same as the phonon frequency distribution of Fig. 4.

More recently Allen⁽³⁾ has noted a relationship between phonon lifetimes due to the electron-phonon interaction and $\alpha^2(\omega)F(\omega)$. The phonon inverse lifetime $\gamma_{\underline{q}\lambda}$ is related to the width of the phonon group and is given by the Fermi Golden Rule

$$\gamma_{\underline{q}} = \frac{2\pi}{\hbar} \sum_{\underline{k}'\underline{k}} |g_{\underline{k}'\underline{k}}| \delta^{2}(\varepsilon_{\underline{k}}) \delta(\varepsilon_{\underline{k}'}) \delta(\underline{k}',\underline{k}+q) \omega_{\underline{q}}$$
(6)

It is easy to see that

$$\alpha^{2}(\omega)F(\omega) = \frac{\hbar}{2\pi N(0)\omega} \begin{cases} F.B.Z. \\ \sum \\ g\lambda \end{cases} \delta(\omega - \omega_{\lambda}(\underline{q})) \qquad (7)$$

where N(0) is the single spin electronic density of states at the Fermi surface

$$N(0) = \sum_{\underline{k}} \delta(\varepsilon_{\underline{k}}) .$$

Butler, Smith and Wakabayashi⁽⁴⁾ have recently presented measurements and calculations of the phonon lifetimes in Nb. Their results are shown in Fig. 8. It is to be noticed that calculations agree well with the available measurements. Because of this good agreements with experiment they extended their calculations to 5200 points in the irreducible wedge of the Brillouin zone and evaluated eq. (7) from this data to get $\alpha^2(\omega)F(\omega)$ which is shown in Fig. 9. It agrees well with the phonon frequency distribution $F(\omega)$.



Fig. 8. Phonon lifetimes in Nb (Butler et al $^{\left(4
ight)}$)



Fig. 9. Calculation of $\alpha^2 F$ from the width of the neutron group (Butler et al(4))

Actually it is possible to measure $\alpha^2(\omega)F(\omega)$ directly in superconducting tunneling experiments. I shall now describe these beautiful and important experiments.

The basic idea is illustrated in Fig. 10. A superconducting film is



Fig. 10

oxidized to get and oxide layer of ~ 20 Å. A normal metal film is then grown on the oxide. The result is a tunnel junction. The oxide layer is to be thick enough that electrons cannot diffuse from one side to the other. Quatum mechanically they can still tunnel. Referring again to Fig. 10 you see a potential barrier representing the oxide layer. On one side of the barrier is shown the wave function of an electron which decays exponentially in the barrier but still leaks out to the other side. There is a finite probability for an electron to tunnel from one side of the oxide layer to the other.

The current (1) flowing through the junction as a function of the voltage drop (V) across it contains a sharp and detailed image of $\alpha^2(\omega) F(\omega)$ and μ^* . This remarkable fact allows us to measure these two parameters and so obtain very valuable microscopic information on particular metals. The exact relationship between I as a function of V and $\alpha^2(\omega)F(\omega)$ is determined by the theory of superconductivity, more specifically through the Eliashberg gap equations.



Fig. 11. The normalized conductance $\sigma(V)$. From McMillan and Rowell $^{(10)}$

The normalized conductance $\sigma(V)$ of a tunnel junction is defined as dI/dV in the superconducting state divided by its value in the normal state. Experimental results on $\sigma(V)$ vs V for a Pb-Pb junction are shown in Fig. 11. As previously stated, the structure exhibited in this figure is an image of the structure in $\alpha^2(\omega)F(\omega)$.

Tunneling results from inversion of the Eliashberg equations in Pb are shown in Fig. 12 and compared with our calculations. The agreement is quite good and shows that the theory of superconductivity is now well understood. It can be used to study normal state parameters like $\alpha^2(\omega)F(\omega)$ and μ^* .

I will now give a brief discussion of two problems of recent interest to me. The question of soft modes in the Al5 compounds and their possible relationshipo to the high values of the critical temperature T_c found in these systems is of considerable interest. For Nb₃Sn, with a T_c of 18 K, Shirane and Axe⁽⁵⁾ have measured the dispersion curves at several temperatures for acoustic phonons along [110] with [110] polarization and have found considerable softening with decreasing temperature.



Their results are reproduced in Fig. 13 in which elastic constant data is also shown. Large decreases in phonon energies as the temperature is reduced are clearly observed. This lead to the speculation that this softening may be responsible for the observed large value of T_c . Simple

BCS type formulas for the relationship between T_c and $\alpha^2(\omega)F(\omega)$ and μ^* leads to the idea that T_c is a rapidly varying function of λ - μ^* where λ is the mass enhancement parameter given by





A shift to lower frequencies in $\alpha^2(\omega)F(\omega)$ at small ω will increase λ significantly because of the 1/ ω weighting. Roughly $\omega = \sqrt{k_s}/M$ with k_s a typical spring constant. A softening of ω means a reduction of k_s which in turn should mean that the lattice is more easily polarizable. Thus T_c should increase.

We can actually test these arguments quantiatively as follows. Shen⁽⁶⁾ has measured $\alpha^2(\omega)F(\omega)$ for Nb₃Sn and his results are given in Fig. 14. A calculation of T_c from the Eliashberg equations based on this spectrum and his suggested μ^* gives a T_c of 18.05 K in agreement with experiment. Next we can get a precise measure of the role played by low frequency phonons in T_c by simply leaving out all modes below a lower cut-off ω_c in $\alpha^2(\omega)F(\omega)$ and repeating the calculations. Results are shown in Table 1. Leaving all phonons below 3.8 meV out of the calculations reduce T_c by only .16 K. This implies that most of the modes measured in the Shirane-Axe⁽⁵⁾ experiment are of no importance to superconductivity. To get a significant effect on T_c, say 5 K, we need to leave out all phonons below 9.8 meV. We conclude therefore that softening of low frequency modes has no direct relationship to the high T_c of Nb₃Sn.



Fig. 14. $\alpha^2 F$ for Nb₃Sn measured by Shen⁽⁶⁾.

While our results are unambiguous we present the following physical arguments that may help understand the underlying physics. Bergmann and Rainer⁽⁷⁾ were the first to introduce the idea of functional derivatives as a means of understanding how various phonon modes affect T_c . The functional derivative of T_c with respect to $\alpha^2(\omega)F(\omega)|\delta T_c/\delta\alpha^2F(\omega)|$ is by definition the change ΔT_c in T_c induced by adding on to

 $\alpha^2(\omega')F(\omega')$ and additional delta function at $\omega' = \omega$ of weight ϵ i.e.

$$\alpha^{2}(\omega')F(\omega') \rightarrow \alpha^{2}(\omega')F(\omega') + \varepsilon\delta(\omega' - \omega)$$
(7)

normalized to ε in the limit $\varepsilon \rightarrow 0$ i.e.

$$\frac{\delta T_{c}}{\delta \alpha^{2} F(\omega)} = \lim_{\epsilon \to 0} \frac{\Delta T_{c}}{\epsilon}$$
(8)

If the functional derivative is large at ω these phonons are more effective than those for which the functional derivative is small. A calculation of $\delta T_c/\delta\alpha^2 F(\omega)$ using the complete Eliashberg equations yields the results given in Fig. 15. We see that the low frequency modes are ineffective in T_c since $\delta T_c/\delta\alpha^2 F(\omega)$ goes to zero as $\omega \neq 0$. The maximum at $\omega \cong 7k_BT_c$ can be understood as follows.



Fig. 15. The functional derivative of $T_{\rm C}$ with respect to $\alpha^2 F$ and $\alpha^2 F$ itself for Nb₃Sn showing that $\omega \sim 7 k_{\rm B} T_{\rm C}$ are the most important frecuencies for high $T_{\rm C}$.

Consider an atom as shown in Fig. 16 which vibrates off equilibrium according to

$$u = A \sin(\omega t)$$
(9)

where u is the excursion, A the amplitude of the simple harmonic motion and t the time with ω the phonon frequency.



The maximum polarization that can be achieved is to pull it off equilibrium to its full amplitude. But for this polarization to be effective in attracting two electrons to each other the first electron should not be farther away than a typical distance in superconductivity (the coherence distance ξ_0) in the time it takes the ion to vibrate to full amplitude. An electron travels with v_F the Fermi velocity. Thus the time T it remains within ξ_0 is ξ_0/v_F = T. For optimum polarization we need

$$\omega T = \frac{\pi}{2}$$
 so $u = A$.

Therefore the optimum phonon frequency is

$$\omega_{\text{op}} = \frac{\pi}{T} = \frac{\pi}{\xi_0} \quad v_F \stackrel{\sim}{=} 7k_B T_c$$

because in B.C.S. theory $\xi_0 \sim v_F / k_B T_c$. This explains why intermediate frequency modes are most effective for superconductivity.

The second problem I wish to discuss is that of phonon lifetimes in a superconductor. As I have said before when a metal becomes superconducting the simple free electron wave function of Fig. 5 does not apply any longer and to pull and electron out of the condensate takes energy \triangle . A phonon decays thorugh the electron-phonon interaction by the creation of a hole particle pair.



Fig. 17. The neutron group sharpens up at low temperatures when the phonon energy becomes smaller than 2Δ . At 6 K the sample is in the superconducting state. At 26 K it goes to the normal state.

In the superconductor at zero temperature no such hole-particle excitations exist with energy less than 2Δ because of the condensation. Thus in a superconductor a phonon with energy less than 2Δ will have an infinite lifetime. The neutron group for that phonon should sharpen up, its width being due only to instrument resolution. This effect is seen clearly in Fig. 17 for Nb₃Sn. The 6 degree results are in a superconducting sample which is normal at 26 K.

Extensive measurements in Nb are summarized in Fig. 18. To understand these results it is necessary to know one more fact about superconductivity. As the temperature is increased real quasiparticle excitations exist in the superconductor. These excitations carry the heat. Their presence interfers with the condensate which is not quite as bound so that the gap at T,∆(T), is reduced over its value at zero temperature (T=0). The reduction in \triangle is small at small T but increases rapidly as T_c is approached, at which point it is zero. The temperature variation 2A(T) is shown as an insert in Fig. 18 where the values of the energy of the three phonon mode investigated is shown as dashed lines labelled A, B and C. A and B are below the value of 2∆ at zero temperature while C is above. For a given phonon energy the width of the phonon group is measured as a function of temperature. At low temperatures this width is zero for both A and B but not for C since it corresponds to an energy larger than the $2\Delta(0)$ value and hence this phonon can decay via the electron-phonon interaction through the creation of a pair of excitations out of the condensate. As the temperature is increased the width of both A and B increases very slightly due to coupling to the few thermal excitations present. But the width increases quite abruptly as the value of twice the gap (at that temperature) becomes equal to the phonon energy, where a new channel for decay opens up due to the creation of real excitations out of the condensate. This rapid increase occurs at lower temperature for B than for A because the phonon energy in B is higher than in A. These are beautiful results which demonstrate the dramatic effect of the presence of the condensate in the superconducting state.



Fig. 18. Dependence of the neutron group width on temperature for Nb.

The same effect can be observed in the ultrasonic attenuation of finite frequency ultrasound. Two types of experiments are conceivable. The frequency of the ultrasound can be varied to sweep through the gap value at some fixed temperature. Alternatively for a fixed ultrasound frequency the temperature can be varied so as to change the gap value. In both cases a sharp increase in the attenuation should be observed at $\omega = 2\Delta(T)$. This was indeed found to be the case in experiments by Liall et al⁽⁸⁾ for the case of aluminum. The rise at 2 Δ (T), however, was found not be as sharp as expected indicating anisotropy in the gap values. In a real material the gap at the Fermi surface needs not be isotoropic but can change with orientation in momentum space. For a Fermi surface with several sheets it can have quite distinct values from one sheet to the other with less variation on a given sheet. We have recently analyzed the ultrasonic attenuation for this case. Results are presented in Fig. 19 in the case of two distinct gaps. It is seen that the attenuation is small for low temperatures and that it increases abruptly when excitations out of the condensate can be used to attenuate the ultrasound wave. The increase in attenuation, however, proceeds in steps, the higher gap coming into play only at higher temperature than the lower one. These steps have not yet been observed but should be there. The effect promises to be a good probe of gap anisotropy.

Lower cut off	0	2.8	3.8	9.8
$T_{c}(K)$	18.05	18.00	17.89	13.05

TADIE

Calculation of ${\tt T}_{\rm C}$ from the Eliashberg equations based on the $\alpha^2 {\tt F}(\omega)$ and μ^* for Nb₃Sn suggested by Shen⁽⁶⁾. We can get a precise measure of the role played by soft phonons by cutting $\alpha^2 F$ at ω_c and using in the calculations of $T_{\rm C}$ only the part of the curve at higher $\omega\,{}^{\rm s}.$ If we leave out of the calculation all the frequencies up to 3.8 we do not get a very significant change in T_C: soft phonons are not very important for superconductivity.



Fig. 19. Ultrasonic attenuation versus temperature for two values of the gap.

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