EFFECT OF SPATIAL DISPERSION OF OPTICAL PHONONS ON INTERBAND MAGNETO-OPTIC ABSORPTION IN SEMICONDUCTORS

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A theory is developed explaining the charge produced in the shape of the oscillation peak of the interband light absorption coefficient by interaction between electrons and optical phonons in crystals with a small ionic-bond fraction. The effect of spatial dispersion of the optical phonons on the shape of the magnetooptical absorption spectrum is investigated. It is found that spatial dispersion is important at magnetic field strengths satisfying the resonance condition $\Omega_c = \omega_0$ (Ω_c is the cyclotron frequency in the conduction band, ω_0 is the limiting frequency of the longitudinal optical phonons). In this case the shape of the magnetooptic spectrum changes qualitatively, depending on the relation between the dimensionless electron-phonon coupling constant η and the dimensionless parameter δ that characterizes the dependence of the optical phonon frequency on the wave vector $\mathbf{q}$ at small values of $\mathbf{q}$. If $\eta/\delta < 1$, there is one oscillation (an electron transition induced by light into the conduction band from the corresponding valence band level onto a level with a Landau quantum number n = 1 is considered), whose shape is related to δ in a simple manner. But if the inequalities $\eta^{2/3} \gg \delta \gg \eta^2$ hold, then instead of one oscillation there will be two with very different shapes; one is less sloping and the other is narrow and high. The shape of the second maximum (which is located at lower light frequencies) depends on δ. By measuring the frequency dependence of the absorption coefficient when $\Omega_c = \omega_0$ one can obtain information regarding the dependence of the optical phonon frequency on $\mathbf{q}$.

THE magnetooptic absorption spectrum of certain semiconductors of the group was investigated recently experimentally and theoretically in magnetic fields such that the cyclotron frequency Ω_c is close to the limiting frequency ω_0 of the longitudinal optical polarization phonon. A splitting of the peak of the light absorption coefficient, corresponding to a direct interband transition to the level n = 1 of the conduction band (n—number of Landau bands) was observed under these conditions in InSb[1]. In the theory of this effect, the magnitude of the splitting of the peaks and the width of one of them were related to the electron-phonon interaction constants[2]. The calculation in[2] was made under the assumption that the dependence of the phonon frequency on the wave vector $\omega(q)$ can be approximated by a constant ω_0. This assumption, which is valid apparently for InSb, is certainly not satisfied for a number of other III-V crystals, where very strong magnetic fields are necessary to satisfy the condition of doubling of the oscillations.

In the present communication we report the results of a theory of the shape of oscillations of the interband absorption coefficient applied in the magnetic field with allowance for the spatial dispersion of the optical phonons, and ascertain the conditions under which this dispersion is significant.

1. FUNDAMENTAL RELATIONS

We consider a cubic crystal in a constant homogeneous magnetic field directed along one of the symmetry axes and satisfying the condition $\Omega_c\tau > 1$, where τ is the lifetime of the given electronic state, and $\Omega_c = eH/m_c c$ (e—electron charge, H—magnetic field, m_c—effective mass of the electron in the conduction band, c—velocity of light in vacuum). We assume that the conduction band (c-band) and the valence band (v-band) are nondegenerate, parabolic, located in the center of the Brillouin zone, and that a direct dipole transition between them is allowed.

We shall assume that the distortion of the electronic states and consequently the form of the magnetooptic oscillations is caused by the interaction of the electron with longitudinal optical phonons, and that the interaction itself is weak, as is the case in crystals with a small fraction of ionic bonding. In addition, we confine ourselves to the case of zero temperature, when the optical branches of the oscillations of the crystal are not excited, and the interaction is effected only in processes in which optical phonons are emitted.

The Hamiltonian of the interaction of the electrons and holes with the phonons is written in the form

$$\mathcal{H} = \sum_{\rho} \sum_{\mathbf{q}} \sum_{\alpha\alpha'} [C_q I_{\alpha\alpha'}(\mathbf{q}) b_{\mathbf{q}} + \text{h.c.}] a_{\rho\alpha}^{+} a_{\rho\alpha'}, \qquad (1)$$

where the sum over ρ denotes summation over the states in the c- and v-bands; $I_{\alpha\alpha'}(\mathbf{q})$—matrix element of the operator $\exp(i\mathbf{q}\cdot\mathbf{r})$, calculated using the wave functions of the electron in the magnetic field; $a_{\rho\alpha}^{+}$ and $a_{\rho\alpha}^{-}$—operators of creation and annihilations of electrons (ρ = c) and holes (ρ = v), $b_{\mathbf{q}}^{+}$ and $b_{\mathbf{q}}$—the same for phonons with wave vector $\mathbf{q}$; $\alpha = (n_\alpha, k_{z\alpha}, k_{x\alpha})$—set of quantum numbers of the electron in the magnetic field directed along the z axis;

$$|C_q|^2 = \frac{4\pi\alpha_0 l_0^3}{V}\left(\frac{\hbar\omega_0}{l_0 q}\right)^2, \quad l_0^2 = \frac{\hbar}{2m_c\omega_0}; \qquad (2)$$

α_0—dimensionless constant of the coupling of the elec-

trons with the optical phonons[3]:

$$\alpha_0 = e^2(2\hbar\omega_0 l_0)^{-1}(\varepsilon_\infty^{-1} - \varepsilon_0^{-1}); \quad (3)$$

ϵ_0 and ϵ_∞—static and high-frequency dielectric constant; V—normalization volume.

The light absorption coefficient $K(\omega)$ is connected with the correction to the single-electron density matrix f_α by the relation

$$K(\omega) = \frac{2\pi}{cn_0}\sigma(\omega) = \frac{2\pi}{cn_0}\frac{e}{m_0}\mathrm{Re}\Big\{\sum_\alpha P^* f_\alpha\Big\}, \quad (4)$$

where m_0—mass of the free electron, P—matrix element of the momentum for the interband transition, n_0—refractive index of light, $\sigma(\omega)$—complex electric conductivity, ω—frequency of light. If the form of the magneto-optical oscillation does not depend on the scattering of the holes (and this takes place when the condition $\Omega_c = \omega_0$ is satisfied in only one of the bands, say the c-band), then f_α is determined by the single-particle retarded Green's function G_r of the electron in the c-band:

$$f_\alpha = \frac{ieP}{2\hbar\omega_0\omega_{cv}V} G_r(\alpha, \omega + \omega_{v\alpha}); \quad (5)$$

$\omega_{cv} = \omega_{c\alpha} - \omega_{v\alpha}$, $\omega_{c\alpha}$ and $\omega_{v\alpha}$—frequencies of the electron in the c- and v-bands:

$$\omega_{c\alpha} = \Omega_c(n_\alpha + 1/2) + \Omega_c k^2, \quad k = \frac{k_z l_H}{\sqrt{2}} \quad l_H = \Big(\frac{c\hbar}{eH}\Big)^{1/2}$$
$$\omega_{v\alpha} = -\omega_g - \Omega_v(n_\alpha + 1/2) - \Omega_v k^2, \quad \Omega_v = eH/m_v c, \quad (6)$$

ω_g—width of forbidden band, and m_v—effective mass of hole.

In the approximation of zero electron density in the conduction band (it is assumed that the v-band is completely filled and the c-band is empty), the electron retarded Green's function G_r coincides with the causal function and is expressed in terms of the mass operator $\Sigma(\alpha, \epsilon)$:

$$G_r(\alpha, \varepsilon) = [\varepsilon - \omega_{c\alpha} - \Sigma(\alpha, \varepsilon) + is]^{-1}, \quad s \to 0, \quad (7)$$

$\Sigma(\alpha, \epsilon)$ can be calculated with the aid of the standard diagram technique.

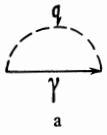
2. DETERMINATION OF THE SPECTRUM OF THE ELECTRON-PHONON SYSTEM

We shall henceforth be interested in the magneto-optical oscillation corresponding to the transition of the electron to the Landau level with n = 1 in the conduction band.

The simplest diagram contained in $\Sigma(\alpha, \epsilon)$ (Fig. 1a) is given by the expression

$$\Sigma_{11} = \sum_{\gamma\mathbf{q}} \frac{\hbar^{-2}|C_q|^2 |I_{\alpha\gamma}(\mathbf{q})|^2}{\varepsilon - \omega_{c\gamma} - \omega(\mathbf{q}) + is}. \quad (8)$$

If the magnetic field is close to resonant, so that $\Omega_c \approx \omega_0$, then the electron projected by the light to the level n = 1 in the region of small values of k_z, will, by virtue of the conservation laws, emit phonons with small q_z, and the $q_\perp$ are limited, as usual, by the condition $q_\perp l_H \lesssim 1$. Therefore we can approximate $\omega(\mathbf{q})$ in the denominator of (8) by a parabola:



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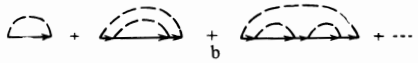
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FIG. 1. Diagrams that must be taken into account in the calculation of the absorption coefficient: a – simplest diagram of phonon emission by an electron, b – renormalization of the simplest diagram.

$$\omega(\mathbf{q}) = \omega_0 \times \Big\{1 - \frac{\Omega_c}{\omega_0}\delta_0\Big[\frac{ql_H}{\sqrt{2}}\Big]^2\Big\}, \quad (9)$$

where the dimensionless parameter is

$$\delta_0 = (2\omega_0)^{-1} l_0^{-2} \times \sum_{i=1}^{3} \frac{\partial^2\omega(0)}{3\partial q_i^2} \ll 1.$$

(We consider below the case $\delta_0 > 0$, which always holds in semiconductors.) Taking (9) into account, Σ_{11} reduces to the form

$$\Sigma_{11} = \frac{\Omega_c\eta}{\pi}\int_0^\infty du \int_{-\infty}^{+\infty} dx \frac{ue^{-u}}{(u + x^2)[\gamma + \lambda - x^2 + \delta u + is]} + \Omega_c\eta(1 + C) - \sum_{n=2}^{\infty}\frac{\Omega_c\eta}{\sqrt{n}}; \quad (10)$$
$$\eta = \frac{\alpha_0}{2}\Big(\frac{\omega_0}{\Omega_c}\Big)^{3/2}, \quad \gamma = \frac{\varepsilon - 3/2\Omega_c}{\Omega_c}$$
$$\lambda = \frac{\Omega_c - \omega_0}{\Omega_c}, \quad \delta = \frac{\Omega_c}{\omega_0}\delta_0,$$

C—Euler's constant.

In order to eliminate the infinite sum in (10), due to the nonresonant terms in the sum over n_γ in (8), it is necessary to renormalize Σ_{11} in a manner shown in Fig. 1b. Denoting the renormalized diagram by Σ_1 and replacing

$$\gamma - \Omega_c\eta C - \Omega_c\eta\sum_{n=2}^{\infty} n^{-1/2}$$

in this diagram by γ, we obtain for Σ_1 (accurate to terms $\sim\delta$):

$$\Sigma_1 = -i\eta\sqrt{\frac{\pi}{\delta}}\exp\Big(\frac{\gamma+\lambda}{\delta}\Big)\Big[1 - \Phi\Big(\sqrt{\frac{\gamma+\lambda}{\delta}}\Big)\Big],$$
$$\Phi(x) = \frac{2}{\sqrt{\pi}}\int_0^x e^{-t^2}dt. \quad (11)$$

The largest of the discarded diagrams (the simplest vertex part) is of the order of smallness η^2/δ.

The spectrum of the electron-phonon system is determined by the roots of the equation with respect to γ:

$$\gamma - k^2 + i\eta\sqrt{\frac{\pi}{\delta}}\exp\Big(\frac{\gamma+\lambda}{\delta}\Big)\Big[1 - \Phi\Big(\sqrt{\frac{\gamma+\lambda}{\delta}}\Big)\Big] = 0 \quad (12)$$

Let us consider the solution of this equation in the limiting case of small k at different relations between the individual parameters.

Let initially $\lambda = 0$ and $\gamma/\delta \lesssim 1$. Then Eq. (12) reduces

to

$$(1-i\theta\sqrt{\pi})\left(\frac{\gamma}{\delta}\right)-2i\theta\left(\frac{\gamma}{\delta}\right)^{1/2}+i\theta\sqrt{\pi}=0,\quad \theta=\frac{\eta}{\delta^{3/2}}, \tag{13}$$

which has two roots:

$$\frac{\gamma}{\delta}=\frac{-i\theta\sqrt{\pi}-(\pi+2)\theta^2\pm\theta\sqrt{4\pi i\theta+(4+\pi)\theta^2}}{1-\pi\theta^2-2i\theta\sqrt{\pi}}. \tag{14}$$

If $\theta \ll 1$, then (14) simplifies and

$$\frac{\gamma}{\delta}=-i\sqrt{\pi}\theta\pm 2^{1/2}\pi^{1/4}(1+i)\theta^{3/2}. \tag{15}$$

It is seen from (15) that the splitting of the levels

$$\Delta\gamma=\mathrm{Re}(\gamma_1-\gamma_2)=2^{3/2}\pi^{1/4}\theta^{3/2}$$

is smaller than their broadening, which equals $-i\sqrt{\pi}\theta$, and consequently in this case two levels of the electron-phonon system (electron in the state n = 1 and electron in the state n = 0 plus one optical phonon) merge into one. If $\theta < 1$ then, as before, the uncertainty of the upper level is larger than $\Delta\gamma$, as can be most readily seen from Fig. 2, which shows plots of $\Delta\gamma$ and Im γ against θ.

In the other limiting case, when $\gamma/\delta \gg 1$, the two aforementioned levels can be resolved, since their width is smaller than the distance between them[2].

Thus, with decreasing parameter η/δ, the distance $\Delta\gamma$ between the two terms of the electron-phonon system decreases from a value $\eta^{2/3}$ ($\delta = 0$) to $2^{3/2}\pi^{1/4}\theta^{3/2}$ ($\theta \ll 1$) and, starting with the values $\theta < 1$, the broadening of the upper level exceeds $\Delta\gamma$. This is connected with a decrease of the number of phonons capable of participating in the transition of the electron from the level n = 1 to the level n = 0. In such a transition, the laws of conservation determine only the z-component of the momentum of the created phonon, and if $\omega(\mathbf{q}) = \omega_0$, then the $q_\perp$ are bounded by the condition $q_\perp l_H \sim 1$. With increasing δ, the law of energy conservation limits $q_\perp$ from above; the larger δ, the greater the limitation. Accordingly, the probability of the transition between the considered Landau bands decreases, together with the distance between terms, which depends on this probability.

From the foregoing results it follows that if $\lambda = 0$ and $\theta \leq 1$, then the spectrum of the system depends essentially on the parameter δ which characterizes the phonon spectrum.

Let now the magnetic field be such that the system is far from resonance: $|\lambda| \gg \gamma$. Since in this case $|\lambda|/\delta \gg 1$, we have in (11):

$$\exp\left(\frac{\lambda+\gamma}{\delta}\right)\left[1-\Phi\left(\sqrt{\frac{\lambda+\gamma}{\delta}}\right)\right] \approx \begin{cases}(\delta/\pi\lambda)^{1/2}, & \lambda>0\\ -i\,(\delta/\pi|\lambda|)^{1/2}, & \lambda<0\end{cases} \tag{16}$$

and Σ_1, together with the form of the spectrum, does not depend on δ.

3. CALCULATION OF THE ABSORPTION COEFFICIENT

The form of the magnetooptical oscillation is determined by the density matrix f_α introduced above, which, when simultaneous account is taken of the scattering of the electrons and the holes, is a solution of an integral equation of the kinetic type[4]. This equation can be iterated with respect to the integral ("arrival") terms, if the scattering in one band (say, in the valence band) is small compared with the scattering in the other. As applied to the situation considered here (for a magneto-optical peak corresponding to transitions to the level n = 1, and satisfaction of the condition $\lambda = 0$ in the c-band), the small parameter of the theory should be taken to be $(m_v\delta/m_c)^{1/2}$ (if $\eta/\delta \ll 1$) or $(m_v/m_c)^{1/2}\eta^{1/3}$ (if $\eta/\delta \gg 1$). These relations were obtained by comparing the simplest diagrams in the non-integral ("departure") terms of the kinetic equation and describing the scattering of the electrons and the holes. All this is valid if $m_v/m_c < 1$, when the hole can also carry out a real transition with emission of an optical phonon. On the other hand, if $m_v/m_c \gg 1$, then the diagram corresponding to "departure" of holes from the given state can become large. However, in this case it is real, since $\Omega_v \ll \omega_0$, and there can be no scattering of a hole with emission of a phonon. But then this diagram is eliminated by renormalization of the width of the forbidden band, and the smallness of the integral terms in the equation will be determined by the parameters $(m_v/m_c)^{1/2}\eta/\sqrt{\delta}$ (if $\eta/\delta \ll 1$) or $\eta^{2/3}\sqrt{m_v/m_c}$ (if $\eta/\delta \gg 1$).

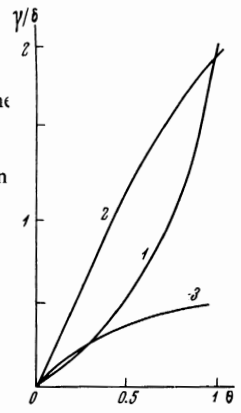

FIG. 2. Dependence of the spectrum of the electron-phonon spectrum on the parameter characterizing the spatial dispersion of the optical phonons. Curve 1 – difference between the upper and lower energy levels, equal to $\mathrm{Re}(\gamma_1 - \gamma_2)$. Curve 2 – sum of the widths of the upper and lower levels, equal to $\mathrm{Im}(-\gamma_1 - \gamma_2)$. Curve 3 – width of lower energy level equal to $\mathrm{Im}(-\gamma_1)$.

We consider below the case most frequently encountered in semiconductors of the III-V type, when $m_c/m_v < 1$. When calculating $K(\omega)$ in this case it is possible to neglect the dependence of the mass operator on k_z, since the form of the peak is determined by small k_z, and in addition, k_z enters in Σ_1 in the combination $k_z^2 m_c/m_v$. Using (4), (5), (7), and (11) we find that the absorption coefficient describing the oscillation n = 1 equals:

$$\begin{aligned}K(\omega)&=K_0\,\mathrm{Re}\{\Gamma+i\eta(\pi/\delta)^{1/2}Q(\sqrt{\zeta})\}^{-1/2},\\ Q(x)&=\exp(x^2)[1-\Phi(x)],\quad x>0,\\ Q(x)&=\exp(-x^2)\left[1-\frac{2i}{\sqrt{\pi}}\int_0^x\exp(t^2)\,dt\right],\quad x<0,\end{aligned} \tag{17}$$

where

$$\Gamma=\frac{\omega-\omega_g-3/2(\Omega_c+\Omega_v)}{\Omega_c},\quad \zeta=\frac{\Gamma+\lambda}{\delta},$$

$$K_0=\frac{\pi}{cn_0}\frac{\sqrt{2}e^2|P|^2m_c}{(2\pi)^2\hbar^2\omega_g m_0^2 l_H}.$$

Let us consider the frequency dependence of $K(\omega)$ at the resonance point ($\lambda = 0$). In the region $|\Gamma|/\delta < 1$, the function $K(\omega)$ takes the form

$$2[K(\omega)/K_0]^2 = (\sqrt{B^2+\pi\eta^2/\delta}+B)/(B^2+\pi\eta^2/\delta), \quad (18)$$

where B = Γ if Γ > 0 and B = − |Γ| + $2\eta\sqrt{|\Gamma|}/\delta$ if Γ < 0.

The absorption in the region of optical frequencies satisfying the condition |Γ|/δ > 1 is described by the formula

$$2\left[\frac{K(\omega)}{K_0}\right]^2 = (\sqrt{\Gamma^2+\eta^2/\Gamma}+\Gamma)/(\Gamma^2+\eta^2/\Gamma), \quad \Gamma>0;$$

$$2\left[\frac{K(\omega)}{K_0}\right]^2 = \left\{\left[\left(-|\Gamma|+\frac{\eta}{\sqrt{|\Gamma|}}\right)^2+\frac{\pi\eta^2}{\delta}e^{-2|\Gamma|/\delta}\right]^{1/2}-|\Gamma| + \frac{\eta}{\sqrt{|\Gamma|}}\right\}\left[\left(-|\Gamma|+\frac{\eta}{\sqrt{|\Gamma|}}\right)^2+\frac{\pi\eta^2}{\delta}e^{-2|\Gamma|/\delta}\right]^{-1}, \quad \Gamma<0. \quad (19)$$

It is seen from (18) that at the points $\Gamma_1 = 0$ and $\Gamma_2 = -4\eta^2/\delta^2$ the values of K(ω) coincide. If the point Γ_2 falls in the region |Γ|/δ < 1 (i.e., if $4\eta^2/\delta^2 < 1$), then a maximum of K(ω) is located between Γ_1 and Γ_2, and there is one oscillation. On the other hand, if η/δ > 1, then formula (18) describes a very narrow interval of the absorption curve in the region of small Γ, and the main part of the oscillation is described by formula (19). In this case the picture changes essentially: at the point Γ_1 there is located a minimum of K(ω), and maxima are located on both sides of this point (at $\Gamma \approx \pm\eta^{2/3}$). These maxima differ greatly in form: the right-hand one (Γ > 0) is more gently sloping and is determined by the parameter η, while the left-hand one (Γ < 0) is narrow and tall, since its form is connected with the exponential term in (19) and is determined by the parameter $\eta^{2/3}/\delta$. If the inequalities

$$\eta^{2/3} \gg \delta \gg \eta^2$$

are satisfied, then the form of these peaks is described (within the framework of the given mechanism of interaction) by the theory, since the largest of the discarded diagrams is $\sim\eta^2/\delta$.

The form of K(ω) for different limiting cases is shown in Fig. 3. In the case when η/δ < 1, the width of the obtained single oscillation is determined from formula (18), and equals

$$\Delta_1 \cong \frac{\sqrt{\pi}\,\eta}{\sqrt{\delta}}\hbar\Omega_c = \frac{\sqrt{\pi}\,\alpha_0}{2\sqrt{\delta}}\hbar\omega_0, \quad (20)$$

while the absorption coefficient at the maximum is

$$[K(\omega)/K_0] = (\delta/4\pi\eta^2)^{1/4}. \quad (21)$$

If the magnetic field is far from resonance (|λ| ~ 1), then there is likewise only one oscillation, but its form ceases to depend on δ, but is determined by the values of η and λ.

Thus, allowance for spatial dispersion of the phonons greatly influences the form of the magnetooptical oscillations, changing qualitatively the form of the spectrum. For example, if η/δ < 1, then we should observe not two closely lying maxima, as is the case in weak dispersion, but only one, whose form is determined by the parameter δ which characterizes the dependence of the frequency of the optical phonons on the wave vector q in the case of small q. The effect of the dependence of the form of the oscillation on the spatial dispersion of the phonons becomes manifest only if the condition $\Omega_c = \omega_0$ is satisfied.

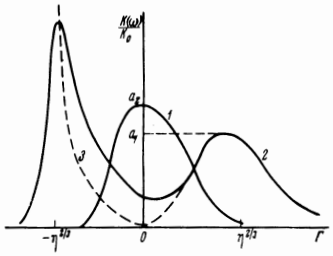

FIG. 3. Frequency dependence of the magnetooptical oscillation. Curve 1 corresponds to the case η/δ < 1, curve 2 – η/δ > 1, $\eta^2/\delta \ll 1$, and curve 3 – to the case δ = 0, $a_1 \approx \eta^{1/3}$, $a_2 = (\delta/4\pi\eta^2)^{1/4}$.

In what kinds of substances can we expect satisfaction of the inequality η/δ < 1, when the influence of the spatial dispersion is most clearly pronounced? Since $\partial^2\omega/\partial q^2 \approx \omega_0 a^2$, where a is the lattice constant, it follows that $\delta_0 \approx (1/2)(a/l_0)^2$. On the other hand, in III-V crystals, the dimensionless coupling constant is $\eta \sim 10^{-2}$. Therefore, satisfaction of the condition η/δ < 1 can be expected in those cases when the optical length l_0 is not too large. Naturally, in this case, in order to obtain the resonance condition $\Omega_c = \omega_0$ (which is equivalent to the condition $2l_0^2/l_H^2 = 1$, strong magnetic fields are necessary. For example, in GaSb $(a/l_0)^2 \approx 6 \times 10^{-2}$, the resonant magnetic field is 3×10^5 Oe, $\eta = 10^{-2}$, and η/δ ≈ 1/3.

Thus, in similar substances we can determine from the form of the magnetooptical oscillations the form of the spectrum of the optical phonons in the case of small momenta.

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