The infrared conductivity of graphene on top of silicon oxide

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Abstract – We study the infrared conductivity of graphene at finite chemical potential and temperature taking into account the effect of phonons and disorder due to charged impurities and unitary scatterers, that is, considering all possible single-particle scattering mechanisms. The screening of the long-range Coulomb potential is treated using the random phase approximation coupled to the coherent potential approximation. The effect of the electron-phonon coupling is studied in second-order perturbation theory. The theory has essentially one free parameter, namely, the number of charge impurities per carbon, $n_F$. Our most important results are the finding of an anomalous enhancement of the conductivity in a frequency region that is blocked by Pauli exclusion, in a picture based on independent electrons, and an impurity broadening of the conductivity threshold, close to twice the chemical potential. We also find that phonons induce Stokes and anti-Stokes lines that produce an excess conductivity, when compared to the far infrared value of $\sigma_0 = (\pi/2)e^2/h$.

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Introduction. – There has been enormous experimental [1] and theoretical [2] progress in understanding the physical properties of graphene since its isolation in 2004 [3]. Among the new phenomena observed in this two-dimensional allotrope of carbon, the transport properties were arguably the most controversial [4–6]. The Kubo formula predicts a universal, impurity-independent, D.C. conductivity: $\sigma_{D.C.,\text{theo}}(\mu = 0) = (4/\pi)e^2/h$ ($\mu$ is the chemical potential measured relative to the Dirac point). Nevertheless, experiments [1] find that $\sigma_{D.C.,\text{exp.}}(\mu = 0) \approx 4e^2/h$ with sample-to-sample variations by a factor of 2, which importantly are in the direction of higher conductivity, i.e., further away from the theoretically predicted value. This discrepancy has been assigned to the macroscopic inhomogeneity and non-local transport in graphene samples [1]. Also the importance of Coulomb impurities for the understanding of the transport properties of single-layer graphene is now well established theoretically [7] and experimentally [8].

In order to gain more insight in the transport and scattering mechanisms in graphene, an aspect of major scientific and technological significance, it is also important to study electronic transport away from the static regime [9–15]. In this regard, the frequency-dependent (A.C.) conductivity, $\sigma(\omega, \mu)$ (we use units such that $\hbar = 1$), provides important information on the several scattering mechanisms of the charge carriers for frequencies $\omega \lesssim 2\mu$. The basic physical processes involved in the A.C. conductivity are easy to understand. A graphene sample is illuminated with light of frequency $\omega$ and vanishing small wave vector that causes creation of particle-hole pairs (pair creation), as shown in fig. 1. At zero temperature and in the absence of disorder or phonons only particle-hole pairs with energy greater than $2\mu$ are allowed, since all the states with energy between $-\mu$ and $+\mu$ are forbidden transitions due to Pauli’s exclusion principle.

In this case, the A.C. conductivity at zero temperature is simply a step function $\sigma(\omega, \mu) = \sigma_0\Theta(\omega - 2\mu)$, where $\sigma_0 = (\pi/2)e^2/h$ is the optical conductivity that has been measured recently [16]. As we shall see, both disorder and phonons will induce spectral weight for frequencies smaller than $2\mu$, a result that can be accounted for by an independent electron theory.

The far-infrared conductivity is insensitive to phonons, impurities (since these affect only the low-energy part of the spectrum) and band structure effects when the frequency of incident light is much larger than $2\mu$ and much smaller than the electronic bandwidth, $W \approx 9\text{eV}$ [17]. Nevertheless, as we are going to show, the infrared spectrum is very sensitive to phonons and...
impurities and the response of the system deviates substantially from the non-interacting clean picture described above. This deviation in turn will illuminate the importance of the several scattering mechanisms present in the system.

Phonons of frequency $\omega_0$ can be either absorbed or emitted by the Dirac electrons. When these phonons are at the center of the Brillouin zone (Γ point) they can be probed by Raman spectroscopy [1], playing an analogous role as light in an A.C. conductivity experiment, that is, creation of particle-hole pairs [18]. Furthermore, impurities play a fundamental role at low energies since it is known that they produce strong broadening of the linewidths [10]. We stress once more, as in the case of D.C. transport, that the impurity broadening has to be calculated self-consistently.

In this paper we compute $\sigma(\omega, \mu)$ taking into account the combined effect of impurities and phonons. We assume that there is a density $n_C^i$ of charge impurities per carbon which might be trapped in the substrate (the silicon oxide), on top of graphene, or in the interface of graphene and the substrate. We model the screening of charge impurities via the random phase approximation (RPA) together with the coherent potential approximation (CPA), which gives us the self-consistent density of states. We also assume a density $n_i$ of unitary scatterers that exist due to structural disorder (edge defects, cracks, vacancies, etc.). The effect of unitary scatterers is only important in producing a finite density of states at the Dirac point and this can be obtained with arbitrarily small values of $n_i$. We assume throughout the paper that impurities are dilute and the structural disorder is very weak, that is, $1 \gg n_C^i \gg n_i \rightarrow 0$.

We have checked that the effect of in-plane acoustic phonons is negligible and they will be ignored in what follows. We assume that the coupling of graphene to the substrate is strong enough to shift the flexural phonon frequencies away from the infrared regime, allowing us to ignore them for the moment being\textsuperscript{1}. Hence, we have kept only the optical phonon modes. The phonon frequency, and the value of the electron-phonon coupling is fixed from Raman experiments [1] and therefore they are not fitting parameters here. In fact, we have only one fitting parameter, namely, $n_i^C$ which will be determined to be $n_i^C \sim 0.5 \times 10^{12}$ cm$^{-2}$, in complete agreement with current experimental estimates [8]. In fact, the importance of impurities in the dispersion of graphene have been appreciated experimentally [19].

**Model Hamiltonian.** – The Hamiltonian has the form:

$$H = H_0 + H_{\text{ph.}} + H_{e-ph.} + H_{\text{imp.}},$$

(1)

where

$$H_0 = -t \sum_{\mathbf{R}, \sigma} \sum_{\delta} \left( a^\dagger_\sigma(\mathbf{R}) b_\sigma(\mathbf{R} + \mathbf{\delta}) + \text{h.c.} \right)$$

(2)

is the nearest-neighbor tight-binding kinetic energy where $a^\dagger_\sigma(\mathbf{R})$ ($b^\dagger_\sigma(\mathbf{R} + \mathbf{\delta})$) creates an electron on site $\mathbf{R}$ of sub-lattice $A (B)$ with spin $\sigma$ ($\sigma = up, \down$), $t \approx 3$ eV is the hopping energy and $\mathbf{\delta}$ are the nearest-neighbor vectors [2].

The phonon Hamiltonian has the form [20–24]:

$$H_{\text{ph.}} = \sum_{\mathbf{R}} \left\{ \frac{P^2_{A}(\mathbf{R})}{2M_C} + \frac{P^2_{B}(\mathbf{R} + \mathbf{\delta}_i)}{2M_C} + \sum_{\delta} \frac{\alpha}{2a^2} \left[ (\mathbf{u}_A(\mathbf{R}) - \mathbf{u}_B(\mathbf{R} + \mathbf{\delta})) \cdot \mathbf{\delta} \right]^2 + \sum_{\delta} \frac{\beta a^2}{2} \left[ \cos(\theta(\mathbf{R}, \mathbf{\delta})) - \cos(\theta_0) \right]^2 \right\},$$

(3)

where $\mathbf{u}_{A,B}$ are the displacements of the $A (B)$ atoms from equilibrium ($\mathbf{P}_{A,B}$ the momentum operator), $M_C$ (= 12 a.u.) is the carbon mass, $\alpha$ (≈ 500 N/m) is the stretching elastic constant, $\theta(\mathbf{R}, \mathbf{\delta}) = \theta_{ij}$ is the angle formed between the $i$-$j$ bond and the $i$-$k$ bond ($\theta_0 = 120^\circ$ is the equilibrium angle) and $\beta$ (≈ 10 N/m) is the in-plane bending elastic constant ($a = 1.42 \text{ Å}$ is the carbon-carbon distance). Although (3) describes both acoustic and optical phonon modes, we focus on the optical modes which can be written as:

$$\mathbf{v}(\mathbf{R}) = (\mathbf{u}_A(\mathbf{R}) - \mathbf{u}_B(\mathbf{R} + \mathbf{\delta}_i))/\sqrt{2},$$

(4)

with frequency $\omega_0^2 = 3(\alpha + \beta/2)/M_C \approx 0.2 \text{ eV} (\approx 1600 \text{ cm}^{-1})$ [21].

\textsuperscript{1}The effect of flexural modes can be analyzed in a similar way but it will introduce new free parameters that have to be adjusted experimentally. Here, we simplify by considering a single adjustable parameter.
The electron-phonon Hamiltonian can be written as
\[
H_{\text{e-ph}} = -\frac{1}{a} \frac{\partial t}{\partial a} \frac{1}{\sqrt{N_c}} \sum_{Q,k,q,\sigma,\delta} \sqrt{\frac{\hbar}{M c_0 \omega_{\nu}(Q)}} \epsilon_{\nu}(Q) \cdot \delta \times (B_{Q,\nu}^\dagger b_{q,\sigma}^{\dagger}(k+Q)b_{q,\sigma}(k) + e^{-ik \cdot \delta} b_{q,\sigma}^{\dagger}(k)a_{\sigma}(k-Q)),
\]
where \( \partial t/\partial a \approx 6.4 \text{eV} \) is the electron-phonon coupling, \( B_{Q,\nu}^\dagger \) creates a phonon of momentum \( Q \), polarization \( \nu \) (polarization vector \( \epsilon_{\nu}(Q) \)), and frequency \( \omega_{\nu}(Q) \) \((N_c \text{ is the number of unit cells})\) [21].

The impurity Hamiltonian has the form
\[
H_{\text{imp}} = \frac{1}{N_c} \sum_{p,q,\sigma} V(q)[a_{\sigma}^{\dagger}(p)a_{\sigma}(p+q) + b_{\sigma}^{\dagger}(p)b_{\sigma}(p+q)].
\]

For a screened impurity of bare charge \( Ze \) the potential \( V(q) \) is given [25] by
\[
V(q) = -\frac{Ze^2}{2\epsilon A_c} e^{-qd} q + \gamma,
\]
where \( \epsilon = 3.9 \) is the SiO\(_2\) relative permittivity, \( d \) is the distance of the impurity to the graphene plane, and \( \gamma = \rho(\mu)c^2/(2A_c) \) is the RPA screening wave vector [2], where \( \rho(\mu) \) is the self-consistent density of states \((A_c = 3\sqrt{3}a^2/2 \text{ is the area of the unit cell})\).

**Self-energies.** – Unitary scatterers (representing cracks, edges, vacancies) are modeled using a local potential \( V(q) = U \) and taking \( U \to \infty \). The effect of a dilute concentration of unitary scatterers can be calculated exactly using the T-matrix, leading to a retarded impurity self-energy of the form [10]
\[
\Sigma_{R}^{\text{unit}}(\omega) = \frac{n_i U/\hbar}{1-U/h \sum_{k} G_0(k,\omega+i0^+)/N_c},
\]
where \( G_0(k,\omega) \) is the free electron Green’s function associated with Hamiltonian (2). In order to account for a self-consistent solution of the scattering problem (the so-called CPA approximation [26]) one has to replace \( G_0(k,\omega) \) with \( G(k,\omega) \) in (8). If the potential \( U \) becomes strong enough, the density of states becomes finite at the Dirac point. In fig. 2 we plot the self-consistent density of states obtained from the solution of the CPA problem for short-range scatterers. It is clear that for small values of \( U \), the density of states, \( \rho(\mu) \), is not very much different from the density of states of clean graphene. On the other hand, the imaginary part of the self-consistent self-energy, \( \hbar \Im \Sigma(E) \), is very much asymmetrical around the Dirac point. Even for values of \( U \approx 100 \text{eV} \), \( \rho(\mu) \) shows a strong particle-hole asymmetry. When \( U \to \infty \) the full particle-hole symmetry is recovered [10].

**Fig. 2:** (Color online) Left: self-consistent density of states obtained by the CPA approximation for different values of \( U \). Right: self-consistent imaginary part of the self-energy due to short-range scatterers. The concentration of impurities is \( n_i = 0.001 \). \( \hbar \Im \Sigma(E) \) is multiplied by 3 for \( U = 10 \text{eV} \).

The self-energy due to charged impurities is calculated in second-order perturbation theory as
\[
\Sigma^C(k,\omega) = 4n_i \sum_p |V(k-p)|^2 G_0(p,\omega_n),
\]
where a term of the form \( n_i V(0) \) was absorbed in the definition of the chemical potential. The self-energy (9) is dependent both on the momentum \( k \) and on the frequency. However, we are interested in the effect of the self-energy for momenta close to the Dirac point \( |q| = \frac{2\pi}{a} (1/3, \sqrt{3}/9) \). Within this approximation, the imaginary part of the retarded self-energy becomes diagonal and momentum independent, reading \((d \approx 0)\)
\[
\Im \Sigma^C(k,\omega) \approx -\frac{Z^2e^4}{4\pi^2 e \sqrt{3}^2} |\omega| \left( \frac{2|\omega|}{3\delta} + \gamma \right)^{-2}.
\]

Notice that the imaginary part of the self-energy behaves like \( |\omega| \) at low frequencies and vanishes as \( 1/|\omega| \) at large frequencies (see fig. 3).

Hence, the electron Green’s function in the presence of impurities is written as \( G^{-1}(k,\omega) = G_0^{-1}(k,\omega) - \Sigma^{\text{unit}}(\omega) - \Sigma^C(k,\omega) \). Notice that the density of states, \( \rho(\omega) = -1/(\pi N_c) \sum_k \Im \Sigma(k,\omega) \), should be computed self-consistently since the screening wave vector \( \gamma \) in (7) and (10) depends on \( \rho(\mu) \).

The self-energy due to electron-phonon interaction is also computed at the Dirac point in second-order perturbation theory
\[
\Sigma^{opt}(K,\omega_n) = -9 \left( \frac{\partial t}{\partial a} \right)^2 \frac{1}{M c_0 \omega_0} \frac{1}{N_c} \sum_{Q} \frac{1}{\beta} \sum_{m} D^0(Q,\omega_n)G(K-Q,\omega_n-i\nu_n),
\]
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where \( D^0(Q, i\nu_n) = 2\omega_0/[(i\nu_n)^2 - (\omega_0)^2] \) is the phonon Green’s function. Notice that \( G(k, i\omega_n) \) is the impurity dressed electronic Green’s function. Due to the exclusion principle, the imaginary part of the electron-phonon self-energy vanishes when \( \mu - \omega_0 < \omega < \mu + \omega_0 \), at \( T = 0 \). At high frequencies the self-energy follows the electronic density of states and is, therefore, linear in \( \omega \), as shown in fig. 3.

Since the electronic self-energy \( \Sigma_{\text{opt}}^\mu(K, i\omega_n) \) has been calculated with the full Green’s function, also the phonon propagator has to be dressed due to its interaction with impurities. The phonon propagator shall be renormalized within the RPA approximation, i.e.,

\[
D_{\nu}^{\text{RPA}}(i\omega_n) = \frac{2\omega_0}{(i\nu_n)^2 - (\omega_0)^2 - 2\omega_0 \Pi_{\nu}(i\omega_n)}. \quad (12)
\]

We obtain for the imaginary part of the retarded phononic self-energy

\[
\Im \Pi_{\nu}^{\text{RPA}}(\omega) = \frac{\hbar}{M_c \omega_{\nu}} \left( \frac{\partial f}{\partial \mu} \right)^2 \frac{18}{\sqrt{3\pi} \ell^2} \int \frac{d\omega'}{2\pi} \Theta(\omega', \omega) \times [f(\omega_1 h - \mu) - f(\omega' h + \omega h - \mu)], \quad (13)
\]

where \( f(x) \) is the Fermi function and \( \Theta(\omega', \omega) \) is a dimensionless function given by

\[
\hbar^2 \Theta(\omega', \omega) = \int_0^D d\epsilon \epsilon A(\epsilon, \omega') A(\epsilon, \omega' + \omega), \quad (14)
\]

where \( A(\epsilon, \omega) \) denotes the spectral function \( A(\epsilon, \omega) = -2\Im G(\epsilon, \omega) \) and \( D \) the band cut-off. It turns out that the phononic self-energy corrections are negligible for typical parameters values.

**Electrical conductivity.** – In the presence of an electromagnetic field the hopping energy changes to

\[
t \rightarrow t e^{i(\epsilon/h)A(t)\delta}. \quad (15)
\]

Expanding the exponential up to second order in the vector potential \( A(t) \) and assuming the electric field to be oriented along the \( x \)-direction, the current operator is obtained from \( j_x = -\partial H/\partial A_x(t) \) leading to \( j_x = j_x^D + A_x(t)j_x^P \). The Kubo formula for the conductivity is given by

\[
\sigma(\omega) = \frac{1}{A_s} \frac{1}{i(\omega + i0^+)} \left[ (j_x^D) + \Lambda_{xx}(\omega + i0^+) \right], \quad (16)
\]

with \( A_s = N_c A_x \) the area of the sample and

\[
\Lambda_{xx}(i\omega_n) = \int_0^\beta d\tau e^{i\omega_n \tau} \langle T_r j_x^P(\tau) j_x^P(0) \rangle, \quad (17)
\]

is the current-current correlation function.

The real part of the D.C. conductivity is given by

\[
\Re \sigma(\mu) = \frac{2e^2}{\pi\hbar} K(\mu), \quad (19)
\]

with \( K(\mu) \) given by

\[
K(\mu) = \frac{1}{2I_a} \sum_{s = \pm 1} \left( \frac{D(sA + D)I_a}{(A + sD)^2 + I_a^2} + A \arctan \frac{A}{I_a} + \frac{I_a}{2A} \left( 2\arctan \frac{A}{I_a} - \arctan \frac{A - D I_a}{I_a} \right) - \arctan \frac{A + D}{I_a} \right), \quad (20)
\]

and

\[
I_a = \hbar \Im \Sigma(\mu). \quad A = \mu - \hbar \Re \Sigma(\mu) \quad (21)
\]

and \( D \) the cut-off bandwidth.

Let us now compute the conductivity as a function of the gate voltage \( V_g \), which relates to the chemical potential as \( V_g \propto \mu^2 \). In fig. 4 we show \( \sigma(\mu) \) as a function of \( V_g \) considering both charged impurities and short-range scatterers (left panel). We note that the conductivity, albeit mostly controlled by charged impurities, still has fingerprints of the finite \( U \) scatterers, due to the asymmetry between the hole (negative \( V_g \)) and particle (positive \( V_g \)) branches. The conductivity follows closely the relation \( \sigma(\mu) \propto V_g \), except close to the Dirac point where its value is controlled by the short-range scattering.

If we suppress the scattering due to charged impurities, which should be the case in suspended graphene, only the scattering due to short-range scatterers survive. In this case the right panel of fig. 4 shows that there is a strong
asymmetry between the hole and the particle branches of the conductivity curve even for a value of $U$ as large as 100 eV. Moreover, the smaller the value of $U$ the larger is the asymmetry in the conductivity curve. We note that asymmetric conductivity curves of suspended graphene were recently measured \[27\].

If the experimental data shows particle-hole symmetry of $\sigma(\mu)$ around the Dirac point, one is forced to consider that the dominant source of scattering is coming from either very strong short-range potentials, \textit{i.e.}, scatterers that are in the unitary limit or due to charged impurities. In the calculations ahead we assume the concentration of unitary scatterers to be much smaller than that of charged impurities. As a consequence, unitary scatterers are only responsible for inducing a small density of states at the Dirac point. From fitting the experimental D.C. curves, we can obtain the impurity density of the Coulomb scatterers.

The finite frequency part of the real part of the conductivity is given by

$$\Re\sigma(\omega) = \frac{2e^2}{\pi h} \int \frac{d\omega'}{\omega} \Theta(\omega', \omega)[f(h\omega' - \mu) - f(h\omega' + h\omega - \mu)],$$

(22)

where $f(x)$ is the Fermi function and $\Theta(\omega', \omega)$ is given by eq. (14). The main features of the conductivity can still be understood from fig. 1. Disorder leads to broadening of the energy levels and a finite density of states at the Dirac point. This implies that the Pauli exclusion is not effective in blocking transitions and hence there is always a finite conductivity even from $\omega < 2\mu$. The spectral weight in the “forbidden” region increases with the increase in the number of impurities. The fact that the imaginary part of the electron-phonon self-energy vanishes for electron energies between $\mu - \omega_0$ and $\mu + \omega_0$ indicates that for $2(\mu - \omega_0) < \omega < 2(\omega_0 + \mu)$ the electron-phonon coupling does not produce any effect in the conductivity. For $\mu < \omega_0$, we expect the appearance of an anti-Stokes line at $\omega_{\text{A.S.}} = 2(\omega_0 + \mu)$ and a Stokes line at $\omega_{\text{S}} = 2(\omega_0 - \mu)$. For $\omega_0 < \mu$ the Stokes line lies inside of the Pauli-blocked region and hence it should be suppressed.

In fig. 5 we plot the infrared conductivity of a graphene sheet in units of the far-infrared conductivity $\sigma_0 = \pi e^2 / (2h)$. The main feature is that the conductivity is finite in the range $0 < h\omega < 2\mu$ and increases as the gate voltage decreases. We choose the concentration of unitary scatterers states in fig. 5 to be one order of magnitude smaller than the one of Coulomb scatterers \[25\], and therefore the conductivity is mainly controlled by phonons and charged impurities. Another feature of the curves in fig. 5 is the large broadening of the inter-band transition edge at $h\omega = 2\mu$. Note that this broadening is not due to temperature but to charged impurities, instead. In fact, the broadening for all values of $V_g$ is larger when the conductivity is controlled by charged impurities. As expected, the coupling to phonons produces an anti-Stokes line centered at $2(\omega_0 + \mu)$. For gate voltages with $\mu < \omega_0$ there appears a Stokes line at $2(\omega_0 - \mu)$. We find, however, that the Stokes line is very sensitive to disorder and is fast suppressed by the inclusion of charge impurities. The optical phonons thus induce a conductivity larger than $\sigma_0$ around these frequencies. This effect is washed out at high temperatures and low frequencies. We also note that for large biases the conductivity in the Pauli-blocked region becomes weakly voltage-dependent.

All these effects are consistent with the recent infrared measurements of graphene on a SiO$_2$ substrate \[28\] which are shown as dashed lines in fig. 5. Notice that there is only one fitting parameter involved which is adjusted by the conductivity curve at zero chemical potential (not shown). Whereas for low gate voltage the agreement is good, there is some spectral weight missing for higher gate voltage ($V > 20$ volts). Nevertheless, all theoretical lines predict lower conductivity such that our model with
only one fitting parameter is consistent with the data. Since we have included in our calculation all possible one-particle scattering mechanisms, the missing weight at large gate voltages can only be attributed to electron-electron interactions in graphene, which become important at this electronic densities. It would be important to repeat the experiment for suspended graphene, avoiding all the interference due to the substrate.

Summary and discussion. – To summarize, we have looked into the effect of a finite-strength potential on the self-consistent density of states of graphene, showing that in the situation where the scattering is dominated by this type of scatterers the D.C. conductivity should not present particle-hole symmetry. Asymmetric D.C. curve is always obtained when the scattering is controlled by charged impurities.

We have also studied the infrared conductivity of graphene at finite chemical potential, generalizing the results of ref. [10]. The calculation includes both the effect of disorder (unitary scatterers and charged impurities) and the effect of phonons. The effect of acoustic phonons is negligible, since it induces an imaginary part of the electronic self-energy that is much smaller than the imaginary part induced by either impurities or optical phonons. We find that optical phonons and charge impurities produce important modifications in the infrared absorption leading to large conductivities in the Pauli-blocked energy region of ω < 2μ, predicted by the independent electron picture. The optical phonons also produce a conductivity larger than σ0 around 2μ < ω ≃ 2(ω0 ± μ) due to Stokes and anti-Stokes processes. It is interesting to note that for frequencies away from the Dirac point the imaginary part of the self-energy due to optical phonons is linear in frequency, a behavior similar to that due to electron-electron interactions in graphene [2]. The most important approximation in our calculations is associated with the fact that we have neglected completely flexural modes, since we assume that they are pinned by the substrate and hence have very high excitation energy, that is, away from the infrared regime. Our results compare well to the experimental data of ref. [28] for low gate voltage, specially if ones takes into consideration that the only free parameter is the density of charged impurities. For gate voltage V > 20 volts, the theoretical curves predict lower conductivity which might be an indication that many-body effects become important in this regime. But to clarify this point more experiments are needed.

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