

Infrared absorption and Raman scattering on coupled plasmon–phonon modes in superlattices

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Abstract

We consider theoretically a superlattice formed by thin conducting layers separated spatially between insulating layers. The dispersion of two coupled phonon-plasmon modes of the system is analyzed by using Maxwell's equations, with the influence of retardation included. Both transmission for the finite plate as well as absorption for the semi-infinite superlattice in the infrared are calculated. Reflectance minima are determined by the longitudinal and transverse phonon frequencies in the insulating layers and by the density-state singularities of the coupled modes. We evaluate also the Raman cross section from the semi-infinite superlattice.

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I. INTRODUCTION

Coupling of collective electron oscillations (plasmons) to optical phonons in polar semiconductors was predicted more than four decades ago [1], experimentally observed using Raman spectroscopy in n -doped GaAs [2] and extensively investigated since then (see, e.g., [3]). Contrary, the interaction of optical phonons with plasmons in the semiconductor superlattices is much less studied. A two-dimensional electron gas (2DEG) created at the interface of two semiconductors has properties which differ drastically from the properties of its three-dimensional counterpart. In particular, the plasmon spectrum of the 2DEG is gapless [4] owing to the long-range nature of the Coulomb interaction of carriers, $\omega^2(k) = v_F^2 \kappa_0 k / 2$, where v_F is the Fermi velocity and κ_0 is the inverse static screening length in the 2DEG. Coupling of two-dimensional plasmons to optical phonons has been considered in Refs. [5, 6] for a single 2DEG layer. The resulting coupling is usually non-resonant since characteristic phonon energies $\sim 30 - 50$ meV are several times larger than typical plasmon energies. Still, hybrid plasmon-optical-phonon modes are of considerable interest in relation to polaronic transport phenomena [7], Raman spectroscopy and infrared optical absorption experiments.

Plasmon excitations in a periodic system of the electron layers have been discussed in a number of theoretical papers [8, 9], disregarding the phonon modes. In the present paper we analyze the coupled phonon-plasmon modes for a superlattice of 2D electron layers sandwiched between insulating layers and demonstrate a possibility of stronger resonant coupling of plasmons to optical phonons excited in the insulator. This enhancement occurs in superlattices due to interaction of plasmons in different layers which spreads plasmon spectrum into a mini-band spanning the energies from zero up to the new characteristic energy, $v_F(\kappa_0/d)^{1/2}$, where d is the interlayer distance [10]. This value could exceed typical phonon frequencies leading to formation of resonant hybrid modes around crossings of phonons and band plasmons.

Usually the coupled phonon-plasmon modes are considered in the so-called electrostatic approximation when the retardation effect is ignored and the terms of ω/c in the Maxwell equations are neglected in comparison with the terms having values of the wave vector k . This is correct if the Raman scattering is studied, when ω has a meaning of the frequency transfer. Then, ω is much less than the incident frequency of the order of $\omega^i \sim ck$. But if we are interested in absorption for the infrared region, when ω is the frequency of incident light

and corresponds to the optical phonon frequency, ω and ck are comparable. In this case, the retardation effect must be fully included.

The plan of the present paper (preliminary results were published in Ref. [11]) is the following. In Sec. II, the Maxwell equations for the periodic system of thin conducting layers sandwiched between the insulating layers are solved to find a spectrum of coupled phonon-plasmon modes. In Sec. III, we consider absorption of the finite sample of the layers and reflectance of the semi-infinite system in the infrared region. In Sec. IV, we analyze the Raman light scattering from the semi-infinite system of layers.

II. SPECTRUM OF COUPLED MODES

We consider a superlattice formed by periodically grown layers of two polar semiconductors (e.g., GaAs and AlGaAs) with 2DEG layers formed in the interface regions, see Fig. 1.

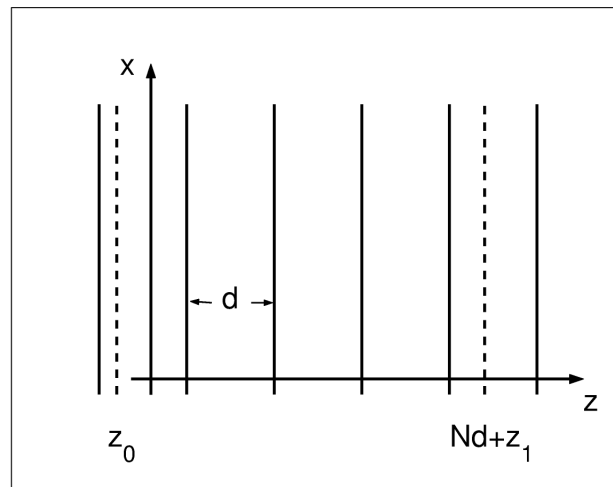


FIG. 1: The stack of 2d conducting layers separated between dielectric layers of thickness d with dielectric constant $\varepsilon(\omega)$; z_0 and $Nd+z_1$ are the boundaries (dashed lines) of the sample if the finite stack is considered.

For the sake of simplicity we assume a superlattice with a single period d and the thickness of a 2DEG layer much less than the period. We also neglect the difference in bulk phonon properties of the two materials. Optical phonons in polar semiconductors are most conveniently described within the dielectric continuum model, which yields the familiar phonon

contribution into the dielectric function,

$$\varepsilon(\omega) = \varepsilon_\infty \frac{\omega_{\text{LO}}^2 - \omega^2 - i\omega\Gamma}{\omega_{\text{TO}}^2 - \omega^2 - i\omega\Gamma},$$

where ω_{LO} and ω_{TO} are the frequencies of longitudinal and transverse optical phonons, respectively, and Γ is the the phonon width (we do not distinguish the widths of the TO and LO modes).

Collective modes of our system are described by the Maxwell equations, which in the Fourier representation with respect to time have the form

$$\nabla(\nabla \cdot \mathbf{E}) - \nabla^2 \mathbf{E} = \varepsilon \frac{\omega^2}{c^2} \mathbf{E} + \frac{4\pi i\omega}{c^2} \mathbf{j}, \quad (1)$$

where the last term takes into account the in-plane electric currents \mathbf{j} induced in the 2DEG layers by the electric field \mathbf{E} . As usual, when the frequency of collective mode lies above the electron-hole continuum, it is sufficient to use the Drude conductivity to describe the in-plane electric currents,

$$\mathbf{j}_\parallel(\omega, x, z) = \frac{ie^2 n_e}{m(\omega + i\gamma)} \sum_n \delta(z - z_n) \mathbf{E}_\parallel(\omega, x, z), \quad (2)$$

where $z_n = nd$ are positions of interfaces (n is integer corresponding to the periodicity of the stack), $n_e = p_F^2/2\pi\hbar^2$ and m are the electron density (per the surface unit) and the effective mass, γ is the electron collision frequency, x and z are the coordinates along and perpendicular to the interfaces, respectively.

We consider the case of the p -polarization, when the field \mathbf{E} lies in the xz -plane and, therefore, the current \mathbf{j} has then only the x -component. Making use of the Fourier transformations with respect to the x -coordinate, $\mathbf{E} \sim e^{ik_x x}$, we can rewrite the Maxwell equations (1) in the form

$$\begin{aligned} ik_x \frac{dE_z}{dz} - \frac{d^2 E_x}{dz^2} - \varepsilon(\omega/c)^2 E_x &= \frac{4\pi i\omega}{c^2} j_x \\ ik_x \frac{dE_x}{dz} + (k_x^2 - \varepsilon\omega^2/c^2) E_z &= 0. \end{aligned}$$

Eliminating

$$E_z = -\frac{ik_x}{\kappa^2} \frac{dE_x}{dz},$$

we get for E_x the equation

$$\left(\frac{d^2}{dz^2} - \kappa^2 - 2\kappa C \sum_n \delta(z - z_n) \right) E_x(\omega, k_x, z) = 0, \quad (3)$$

where

$$C = \frac{2\pi n_e e^2 \kappa}{\varepsilon \omega (\omega + i\gamma) m}, \quad \kappa = \sqrt{k_x^2 - \varepsilon \omega^2 / c^2}.$$

At the interfaces $z = z_n$, the E_x component must be continuous and the z -component of electric induction εE_z has a jump:

$$\varepsilon(E_z|_{z=nd+0} - E_z|_{z=nd-0}) = 4\pi \int_{nd-0}^{nd+0} \rho(\omega, k_x, z) dz, \quad (4)$$

where the carrier density is connected to the current (2) according to the equation of continuity

$$\rho(\omega, k_x, z) = j_x(\omega, k_x, z) k_x / \omega. \quad (5)$$

For the infinite stack of layers, $-\infty < n < \infty$, independent solutions to Eqs. (3) – (4) represent two Bloch states $E_x(z) = f_{\pm}(z)$ moving along positive and negative directions of the z -axis,

$$f_{\pm}(z) = e^{\pm i k_z n d} \sinh \kappa(z - n d) - e^{\mp i k_z d} \sinh \kappa[z - (n + 1)d], \quad n d < z < (n + 1)d \quad (6)$$

with the quasi-momentum k_z determined from the dispersion equation:

$$\cos k_z d = \cosh \kappa d - C \sinh \kappa d. \quad (7)$$

The quasi-momentum k_z can be restricted to the Brillouin half-zone $0 < k_z < \pi/d$, if the phonon and electron damping vanishes. In the general case, we fix the choice of the eigenfunctions in Eq. (6) by the condition $\text{Im} k_z > 0$, so that the solution f_+ decreases in the positive direction z . The equation (7) implicitly determines the spectrum $\omega_{\pm}(k_x, k_z)$ of two coupled plasmon–optical-phonon modes shown in Figs. 2-3 as functions of the in-plane wave vector k_x and the quasi-momentum k_z . These modes are undamped if the electron collision rate and the phonon width are small. The modes arise from the interaction of the plasmon branch in the 2DEG and the phonon LO mode in the 3-d insulator. They have a definite character far from the intersection of the corresponding dispersion curves. For instance, at small values of k_x , the $\omega_-(k_x, k_z)$ mode is mainly the plasmon mode, whereas the $\omega_+(k_x, k_z)$ mode has mainly the plasmon character. At large values of k_x , they interchange their character.

Note, that $k_x = k_z = 0$ is a saddle point for both branches $\omega_{\pm}(k_x, k_z)$. In the vicinity of this point, the frequency grows as a function of k_x and decreases with increasing k_z . This

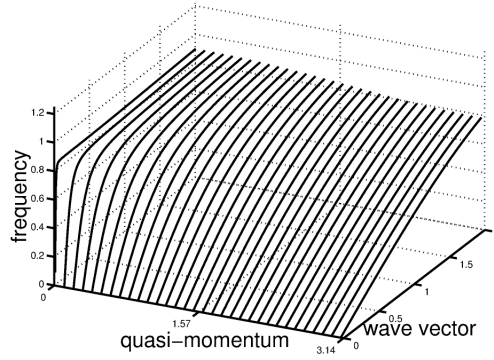


FIG. 2: The plasmon-like (at small wave vectors) mode of an infinite number of 2D metallic layers (with the carrier concentration in every layer $n_e = 3 \cdot 10^{11} \text{ cm}^{-2}$) sandwiched between dielectric layers of thickness d with the phonon frequency ω_{LO} . The frequency $\omega_-(k_x, k_z)$ in units of ω_{LO} is plotted as a function of the in-plane wave vector k_x and the quasi-momentum $k_z < \pi/d$ (both in units $1/d$). Parameters used here are reported in the literature for GaAs: $\omega_{\text{LO}} = 36.5 \text{ meV}$, $\omega_{\text{TO}} = 33.6 \text{ meV}$, $\varepsilon_\infty = 10.6$; the thickness d is taken as $d = 1/\kappa_0$, $\kappa_0 = 2me^2/\hbar^2\varepsilon_\infty = 2.5 \cdot 10^6 \text{ cm}^{-1}$ is the screening length.

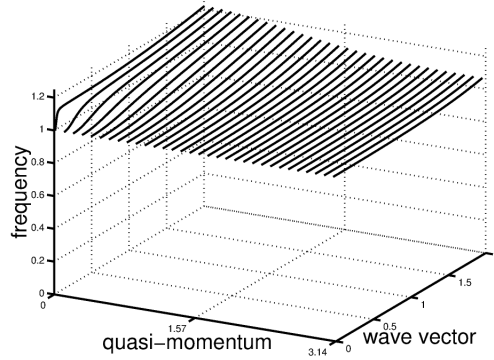


FIG. 3: Same as in Fig. 2 but for the phonon-like (at small wave vectors) mode $\omega_+(k_x, k_z)$.

is evident in Figs. 4-5 and can be explicitly shown in the limit of $k_x \gg \omega|\varepsilon(\omega)|/c$, when the retardation effects of the electromagnetic field are negligible. Then, $\kappa \approx k_x$ and equation (7) yields two solutions,

$$\omega_\pm^2(k_x, k_z) = \frac{1}{2}(\Omega^2 + \omega_{\text{LO}}^2) \pm \frac{1}{2}[(\Omega^2 + \omega_{\text{LO}}^2)^2 - 4\Omega^2\omega_{\text{TO}}^2]^{1/2}, \quad (8)$$

where we introduced the notations,

$$\Omega^2(k_x, k_z) = \omega_0^2(k_x) \frac{\sinh k_x d}{\cosh k_x d - \cos k_z d}, \quad \omega_0^2(k_x) = \frac{2\pi n_e e^2 k_x}{m\varepsilon_\infty}.$$

The frequency $\omega_0(k_x)$ is the conventional square-root spectrum of two-dimensional plasmons in the limit of layer separation large compared to the wavelength, $d \gg k_x^{-1}$. The quantity $\Omega(k_x, k_z)$ describes the plasmon spectrum that would exist in a superlattice of consisting non-polar semiconductors (when $\omega_{LO} = \omega_{TO}$).

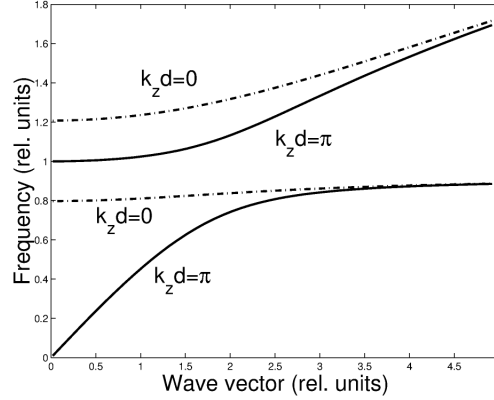


FIG. 4: Dispersion of coupled phonon-plasmon modes of an infinite number of 2D metallic layers sandwiched between dielectric layers. The frequencies $\omega_{\pm}(k_x, k_z)$ (in units of ω_{LO}) are plotted as functions of the in-plane wave vector k_x for two values of the quasi-momentum k_z (all in units of the inverse period $1/d = 2.5 \cdot 10^6 \text{ cm}^{-1}$). The dashed-dotted lines represent the upper boundary $\omega_{\pm}(k_x, k_z = 0)$ of two phonon-plasmon modes, while the solid lines mark the lower boundary corresponding to $\omega_{\pm}(k_x, k_z = \pi/d)$. Parameters are the same as in Figs. 2 - 3.

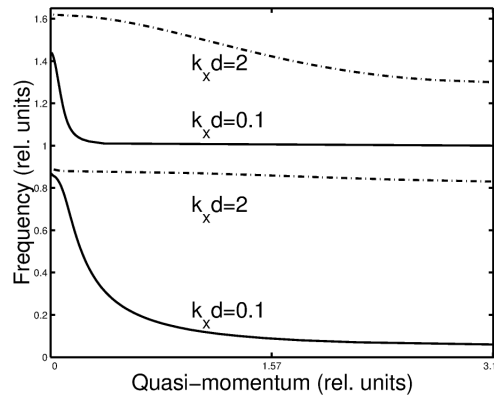


FIG. 5: Dispersion of coupled phonon-plasmon modes. The frequencies $\omega_{\pm}(k_x, k_z)$ are plotted as a function of the quasi-momentum k_z for two values of the in-plane wave vector k_x .

The mode $\omega_+(k_x, k_z)$ has a gap while the other mode has a linear dispersion $\omega_-(k_x, k_z) =$

$s(k_z)|k_x|$ at $k_x \ll (d^{-1}, k_z)$ with the velocity given by

$$s(k_z) = v_F \sqrt{\frac{\kappa_0 d}{2(1 - \cos k_z d)}},$$

where $\kappa_0 = 2me^2/\hbar^2\varepsilon_\infty$ is the static screening radius in the 2DEG. For a fixed value of the in-plane wave vector k_x , every mode develops a band (with respect to the quasi-momentum k_z) with the boundaries, $\omega_{upper}(k_x) \leq \omega(k_x, k_z) \leq \omega_{lower}(k_x)$ where

$$\omega_{upper}^2 = \Omega^2(k_x, 0) = \omega_0^2(k_x) \coth \frac{k_x d}{2}$$

for the upper boundary and

$$\omega_{lower}^2 = \Omega^2(k_x, \pi/d) = \omega_0^2(k_x) \tanh \frac{k_x d}{2}$$

for the lower boundary. Fig. 5 illustrates the behavior of the gapless, so called "acoustic" mode for the small values of $k_z < k_x$, where it acquires a gap. We see that the frequency of this mode decreases rapidly in the region $k_z > k_x$.

In the rest of the paper we analyze various experimental implications resulting from the existence of hybrid plasmon-phonon modes. Such modes can be observed in both the infrared absorption and the Raman spectroscopy.

III. INFRARED ABSORPTION ON COUPLED PLASMON-PHONON MODES

We calculate now the reflectance and the transmission of a plane wave with the p -polarization, incident from the vacuum on the thin plate consisting of the system of layers. Let us suppose that the boundaries of sample are parallel to the layers and intersect the z -axis at $z = z_0$ and $z = Nd + z_1$, respectively, with $0 < z_0, z_1 < d$ (see Fig. 1). We assume in the vacuum ($z < z_0$),

$$E_x(z) = e^{ik_z^{(i)}(z-z_0)} + A e^{-ik_z^{(i)}(z-z_0)},$$

where $k_z^{(i)} = \sqrt{(\omega/c)^2 - k_x^2}$ and A is the amplitude of the reflected wave. In the region $z > Nd + z_1$, the transmitted wave has the form

$$E_x(z) = T e^{ik_z^{(i)}(z-z_1-Nd)},$$

and we search inside the plate the field as a sum of two solutions (6):

$$E_x(z) = C_+ f_+(z) + C_- f_-(z),$$

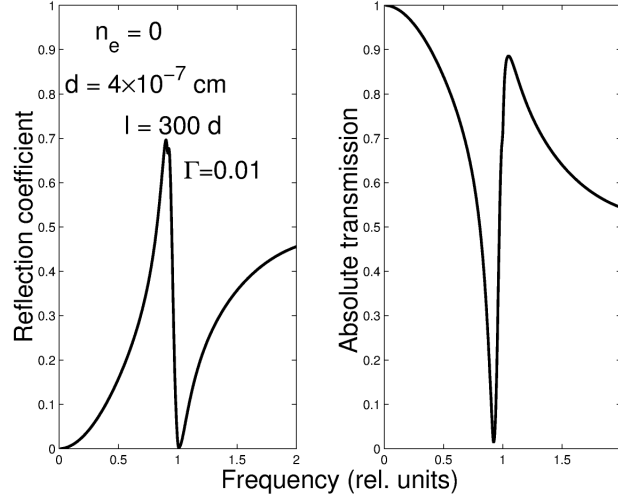


FIG. 6: – Fig. 9: Calculated p-polarized reflection-absorption spectra of GaAs plates (of the thickness $l = 300d$) with the super-lattices of different electron concentrations in a layer; the frequencies, the phonon width Γ , and the electron relaxation frequency γ are given in units of ω_{LO} . The lattice period $d = 1/\kappa_0$ and the incidence angle $\theta = \pi/4$. Parameters used here are reported in the literature for GaAs: $\omega_{LO} = 36.5$ meV, $\omega_{TO} = 33.6$ meV, $\varepsilon_\infty = 10.6$.

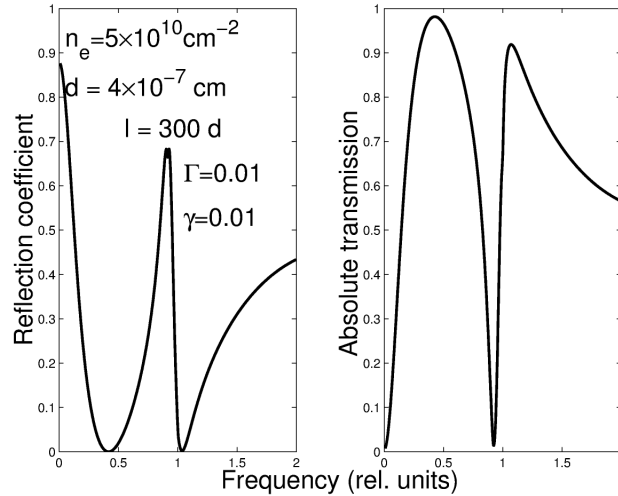


FIG. 7:

with the definite values of k_x , ω , and $k_z = k_z(\omega, \kappa)$ obeying the dispersion equation (7).

As usual, the boundary conditions at $z = z_0$ and $z = Nd + z_1$ require the continuance of the x -component of the electric field E_x parallel to the layers and of the z -component of the electric induction D_z normal to the layers. The second condition rewritten via the

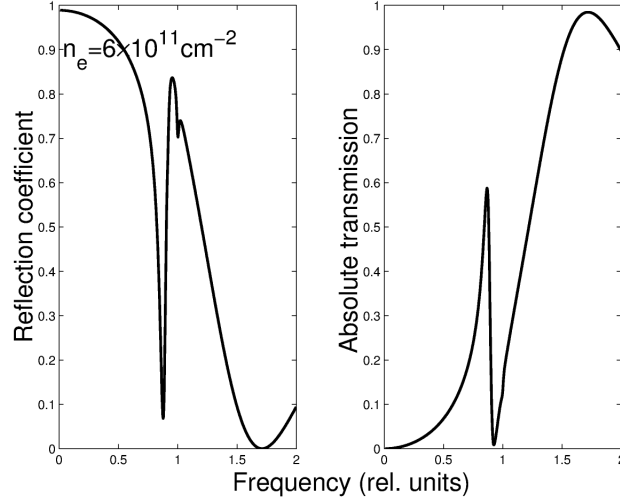


FIG. 8:

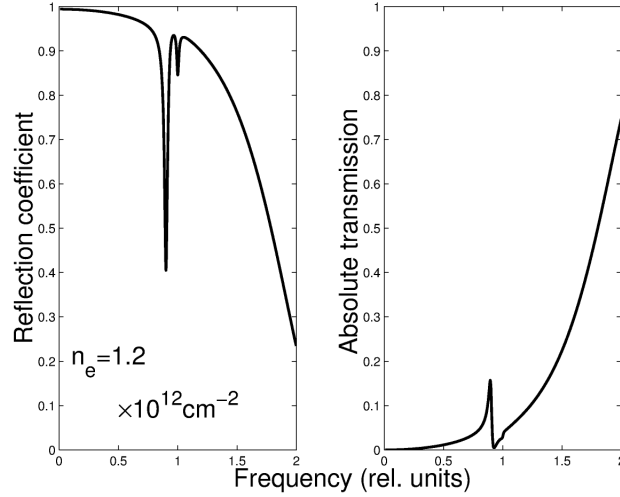


FIG. 9:

electric field E_x gives, e.g., at $z = z_0$

$$\frac{\varepsilon}{\kappa^2} E'_x(z_0+) = -\frac{1}{k_z^{(i)2}} E'_x(z_0-).$$

The boundary conditions give for C_+ , C_- , A , and T the following equations

$$\begin{aligned} 1 + A &= C_+ f_+(z_0) + C_- f_-(z_0), \\ -1 + A &= [C_+ f'_+(z_0) + C_- f'_-(z_0)] \frac{\varepsilon k_z^{(i)}}{i\kappa^2}, \\ T &= C_+ f_+(z_1) + C_- f_-(z_1), \\ -T &= [C_+ f'_+(z_1) + C_- f'_-(z_1)] \frac{\varepsilon k_z^{(i)}}{i\kappa^2}. \end{aligned}$$

Solving these equations, we find the transmitted amplitude

$$T = \frac{2\varepsilon k_z^{(i)} f'_+(z_1)f_-(z_1) - f_+(z_1)f'_-(z_1)}{i\kappa^2 (a_{11}a_{22} - a_{12}a_{21})}$$

and the reflected amplitude

$$A = -1 + 2 \frac{a_{21}f_-(z_0) - a_{22}f_+(z_0)}{a_{11}a_{22} - a_{12}a_{21}},$$

where

$$a_{11} = f'_+(z_0)\varepsilon k_z^{(i)}/i\kappa^2 - f_+(z_0), a_{22} = f'_-(z_1)\varepsilon k_z^{(i)}/i\kappa^2 + f_-(z_1),$$

$$a_{12} = f'_-(z_0)\varepsilon k_z^{(i)}/i\kappa^2 - f_-(z_0), a_{21} = f'_+(z_1)\varepsilon k_z^{(i)}/i\kappa^2 + f_+(z_1).$$

The transmission $|T|^2$ and reflection $|A|^2$ coefficients are shown in Figs. 6-9 for samples of various electron concentrations n_e . The incidence angle is taken $\theta = \pi/4$. Other parameters are the following: the thickness $l = 300 d$, the lattice period $d = 1/\kappa_0 = 4 \cdot 10^{-7}$ cm, the phonon width $\Gamma = 0.02\omega_{\text{LO}}$, and the electron scattering rate $\gamma = 0.01\omega_{\text{LO}}$. In all that case $k_z d \ll 1$, and there are many layers on the wave length of the field. Therefore, the reflectance is not sensitive to the positions of the sample surface z_0 and z_1 . But, if the thickness d is more larger, as shown in Fig. 10, an interference phenomenon is seen.

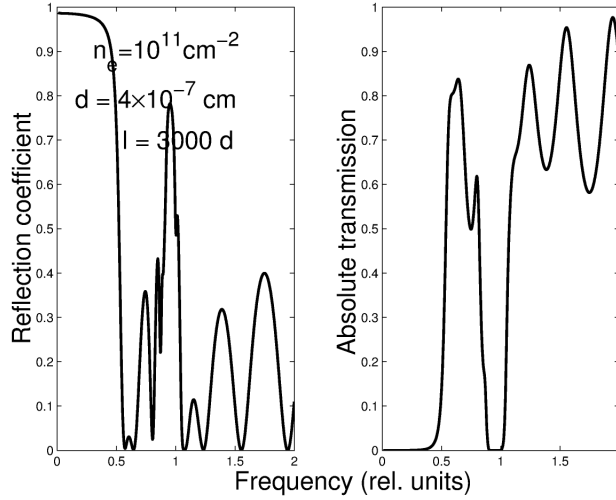


FIG. 10: Interference in the sample of thickness $l = 3000d$; the electron concentration is 10^{11} cm^{-2} .

In order to avoid the interference effect, we calculate the reflectance for the semi-infinite sample. The results can be seen in Figs. 11 - 12, where the theoretical curves are presented

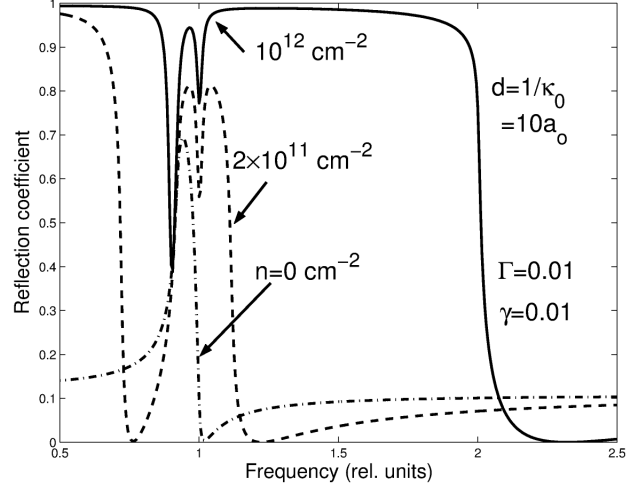


FIG. 11: Calculated p-polarized reflection-absorption spectra for the semi-infinite superlattice via the frequency in units of ω_{LO} at the incident angle $\pi/4$. The electron concentration is labelled on curves, the lattice period is $d = 1/\kappa_0 = 4 \times 10^{-7}$ cm; the phonon width is Γ and the electron relaxation rate is γ (in uns. ω_{LO}).

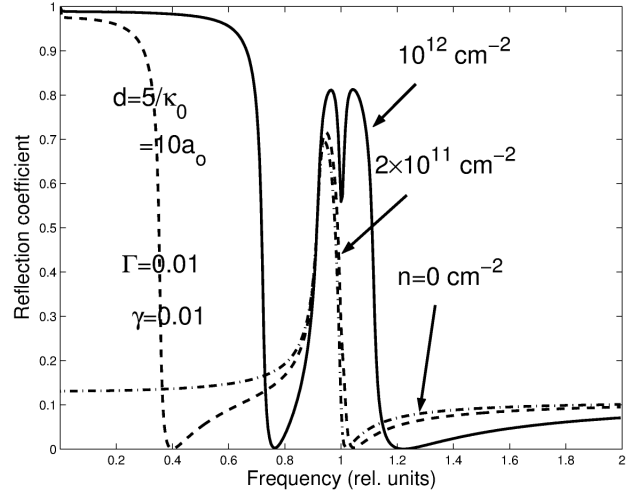


FIG. 12: Same as in Fig. 11, but for larger period.

for the various electron concentrations and the lattice period. There is a singularity at ω_{LO} . For $d = 1/\kappa_0$ and the intermediate electron concentration in the layer (dashed line in Figs. 11-12), there are two regions (one is at higher frequencies and another is just below $\omega_{TO} = 0.9\omega_{LO}$) where the sample with layers is more transparent than the sample without any (shown in the dashed-dotted line).

This is an effect of the coupled phonon-plasmon modes: the minima of the reflection

coefficient correspond to the density state singularities of the phonon-plasmon coupled modes at $k_z = 0$ and $k_x \rightarrow 0$ (see Fig. 4). The frequencies of these singularities are determined by Eq. (8) with $\Omega^2 = 4\pi n_e e^2 / md\varepsilon_\infty$. For the large electron concentration (upper solid line), the reflection is not complete only in the narrow interval bounded by the singularities at ω_{TO} and ω_{LO} . Finally, the reflection coefficient tends to the unity at the low frequencies because the skin depth of the metallic system goes to the infinity in this case. For the sample with the large period $d = 5/\kappa_0$ (Fig. 12), the effect of carriers is seen at the higher concentration.

IV. RAMAN SCATTERING FROM COUPLED MODES

Consider now the Raman scattering of the radiation incident from a vacuum with the vector potential \mathbf{A}^i , the frequency ω^i , and the wave vector \mathbf{k}^i on the sample occupying the semi-infinite space $z > z_0$, where $0 < z_0 < d$. The corresponding quantities in the scattered wave are designated as \mathbf{A}^s , ω^s , and \mathbf{k}^s .

In addition to these fields, an electric field \mathbf{E} is excited in the Raman light scattering in polar crystals along with the longitudinal optical vibrations \mathbf{u} . The field \mathbf{E} corresponds to the excitation of plasmons, whereas the vibrations \mathbf{u} associate with the phonon excitations.

These processes can be described with the help of the effective Hamiltonian

$$\mathcal{H} = \int d^3r \mathcal{N}_{jk}(t, \mathbf{r}) A_j^s(t, \mathbf{r}) A_k^i(t, \mathbf{r}), \quad (9)$$

where the operator

$$\mathcal{N}_{jk}(t, \mathbf{r}) = g_{ijk}^u \hat{u}_i(t, \mathbf{r}) + g_{ijk}^E E_i(t, \mathbf{r}) \quad (10)$$

is linear in the phonon \mathbf{u} and photon operators \mathbf{E} .

We are interested in the inelastic scattering on the phonon-plasmon coupled modes. Therefore, we assume that the frequency transfer $\omega = \omega^i - \omega^s$ is of the order of the phonon frequencies ω_{LO} , but the frequencies ω^i and ω^s of the incident and scattered fields are much greater than the phonon frequencies. Then, we can ignore the effect of carriers in the layers on the propagation of both scattered $\mathbf{A}^s(t, \mathbf{r})$ as well as incident $\mathbf{A}^i(t, \mathbf{r})$ lights, taking the incident field in the sample ($z > z_0$) in the form

$$A_j^i(\omega^i, k_x^i, z) = t_j e^{ik_z^i(z-z_0)},$$

normalized to the incident flow; t_j is the transmission coefficient from vacuum into the sample, $k_z^i = \sqrt{\varepsilon(\omega^i)(\omega^i/c)^2 - k_x^i{}^2}$.

When the scattered field \mathbf{A}^s is taken as the variable in the Hamiltonian (9), we obtain an additional current $\mathbf{j}^s(t, \mathbf{r})$

$$-c \frac{\delta \mathcal{H}}{\delta \mathbf{A}_j^s} = j_j^s(t, \mathbf{r}) = -c \mathcal{N}_{jk}(t, \mathbf{r}) A_k^i(t, \mathbf{r}) \quad (11)$$

in the Maxwell equation for this field. The phonon and plasmon fields in $\mathcal{N}_{jk}(t, \mathbf{r})$ are the source of the Raman scattering, whereas the incident field $\mathbf{A}^i(t, \mathbf{r})$ is considered as the external force for scattering.

Eliminating the z -component, we write the Maxwell equation for the p -polarization of the scattered field ($x - z$ is the scattering plane) in the form

$$\left(\frac{d^2}{dz^2} + k_z^{s2}(\omega^s) \right) E_x^s(\omega^s, k_x^s, z) = I(\omega^s, k_x^s, z),$$

where

$$I(\omega^s, k_x^s, z) = \frac{-4\pi i}{\omega^s \varepsilon(\omega^s)} \left(k_z^{s2}(\omega^s) j_x^s(\omega^s, k_x^s, z) + ik_x^s \frac{dj_z^s(\omega^s, k_x^s, z)}{dz} \right) \quad (12)$$

and $k_z^s(\omega^s) = \sqrt{\varepsilon(\omega^s)(\omega^s/c)^2 - k_x^{s2}}$ is the normal component of the wave vector in the medium for the scattered wave.

The scattered field in the vacuum $z < z_0$ is expressed in terms of $I(\omega^s, k_x^s, z)$:

$$E_x^s(\omega^s, k_x^s, z) = \frac{2\varepsilon(\omega^s)\omega^s \cos \theta^s / c}{k_z^s + \varepsilon(\omega^s)\omega^s \cos \theta^s / c} I_0 e^{-i(z-z_0)\omega^s \cos \theta^s},$$

$$E_z^s(\omega^s, k_x^s, z) = (k^s \omega^s \cos \theta^s / ck_z^{s2}) E_x^s(\omega^s, k_x^s, z),$$

where

$$I_0 = \frac{i}{2k_z^s} \int_{z_0}^{\infty} dz' e^{ik_z^s(z'-z_0) \cos \theta^s / c} I(\omega^s, k_x^s, z') \quad (13)$$

and θ^s is the propagation angle of scattered wave in the vacuum.

The energy flow from the surface of the sample is given by $|E_x^s(\omega_s, k_x^s, z < z_0)|^2$. Therefore, we have to calculate

$$\langle I^*(\omega^s, k_x^s, z) I(\omega^s, k_x^s, z') \rangle \quad (14)$$

averaged quantum-mechanically and statistically, where according to Eqs. (12) and (11) we meet the Fourier transform of the correlation function

$$K_{jk, j'k'}(t, \mathbf{r}; t', \mathbf{r}') = \langle \mathcal{N}_{jk}^*(t, \mathbf{r}) \mathcal{N}_{j'k'}(t', \mathbf{r}') \rangle .$$

Because this correlator depends on the differences $(t - t')$ and $(x - x')$, we can expand that in the Fourier integral with respect to these differences. Then, we have the Fourier transform $K_{jk,j'k'}(\omega, k_x, z, z')$ which can be expressed in terms of the generalized susceptibility according to the fluctuation-dissipation theorem:

$$K_{jk,j'k'}(\omega, k_x, z, z') = \frac{2}{1 - \exp(-\omega/T)} \text{Im} \chi_{jk,j'k'}(\omega, k_x, z, z').$$

The generalized susceptibility $\chi_{jk,j'k'}(\omega, k_x, z, z')$ is involved in the response

$$\langle \mathcal{N}_{jk}(\omega, k_x, z) \rangle = - \int_{z_0}^{\infty} dz' \chi_{jk,j'k'}(\omega, k_x, z, z') U_{j'k'}(\omega, k_x, z') \quad (15)$$

to the force

$$U_{jk}(\omega = \omega^i - \omega^s, k_x = k_x^i - k_x^s, z) = A_j^s(\omega^s, k_x^s, z) A_k^i(\omega^i, k_x^i, z) \sim e^{iq_z z},$$

where $q_z = k_z^i + k_z^s$.

To calculate the generalized susceptibility $\chi_{jk,j'k'}(\omega, k_x, z, z')$, we write the equations for the averaged phonon \mathbf{u} and plasmon \mathbf{E} fields. The variation of the Hamiltonian (9) with respect to the vibrations \mathbf{u} gives in the right-hand side of the motion equation for the phonon field

$$(\omega_{\text{TO}}^2 - \omega^2 - i\omega\Gamma)u_i(\omega, k_x, z) = \frac{Z}{\rho} E_i(\omega, k_x, z) - \frac{g_{ijk}^u}{\rho} U_{jk}(\omega, k_x, z), \quad (16)$$

the additional term to the force from the electric field; ρ is the density of the reduced mass and Z is the effective charge.

The equation for the plasmon field \mathbf{E} can be obtained if this field is taken as the variable in the Hamiltonian (9):

$$\nabla \cdot (\mathbf{E} + 4\pi\mathbf{P}) = 4\pi\rho. \quad (17)$$

The charge density ρ is connected to the current by Eq. (5). The polarization \mathbf{P} includes the dipole moment $Z\mathbf{u}$, the contribution of the filled electronic state $\chi_{\infty}\mathbf{E}$, and the variational term $-\delta\mathcal{H}/\delta\mathbf{E}$:

$$P_i = Zu_i + \chi_{\infty}E_i - g_{ijk}^E U_{jk}.$$

Here we can substitute u_i using Eq. (16). We obtain in Eq. (17) the term with $\varepsilon_{\infty} = 1 + 4\pi\chi_{\infty}$ and $g_{ijk}^E \rightarrow \tilde{g}_{ijk}^E$.

$$\tilde{g}_{ijk}^E = g_{ijk}^E + g_{ijk}^u Z / \rho (\omega_{\text{TO}}^2 - \omega^2 - i\omega\Gamma). \quad (18)$$

Equation (17) can be substantially simplified in the case under consideration when the wave vectors \mathbf{k}^i , \mathbf{k}^s , and consequently the momentum transfer $\mathbf{k} = \mathbf{k}^i - \mathbf{k}^s$ are determined by the frequency of the incident optical radiation, whereas the frequency transfer ω of the excited fields \mathbf{E} and \mathbf{u} is much less than the incident frequency, $\omega = \omega^i - \omega^s \ll \omega^i$. Therefore, we can neglect here the retardation terms ω/c compared to k_x (so that $\kappa = k_x$) and introduce the potential, $\mathbf{E} = -\nabla\phi$. We obtain for the potential the following equation

$$\left(\frac{d^2}{dz^2} - k_x^2 - 2k_x C \sum \delta(z - z_l)\right) \phi(\omega, k_x, z) = \frac{-4\pi}{\varepsilon(\omega)} \left(ik_x \tilde{g}_{xjk}^E + \tilde{g}_{zjk}^E \frac{d}{dz}\right) U_{jk}(\omega, k_x, z). \quad (19)$$

The solution to this equation is found by using the Green's function obeying the same Eq. (19) but with the $\delta(z - z')$ function on the right-hand side. As very well known, the Green's function is expressed in terms of the solutions of the corresponding homogeneous equation. Taking into account $f_{\pm}(\omega, k_x, z)$ (6), we write:

$$G(z, z') = \frac{i}{2\kappa \sin(k_z d) \sinh(\kappa d)} \begin{cases} f_+(z)f_-(z'), & z > z' \\ f_-(z)f_+(z'), & z < z' \end{cases} \quad (20)$$

where where the quasi-momentum $k_z = k_z(\omega^s, \kappa)$ has to be determined from the dispersion relation (7) ignoring the retardation, $\kappa = k_x$.

According to T_d symmetry of GaAs lattice, the Raman tensors g_{ijk}^u and g_{ijk}^E have only two independent components g_{xxx} and g_{xyz} in the crystal axes. Let the scattered light is always polarized in the xz -plane. Now we consider two geometry. For the parallel scattering geometry (a), the incident light considered to be polarized in the x - direction, then the x -components of exited phonon and plasmon fields can be active in the Raman scattering due to g_{xxx} . For the crossed geometry (b), the incident light is polarized in the y - direction. Then, z -components of the exited fields are active and we take into account terms with g_{xyz} . Thus, for the generalized susceptibility $\chi_{xx,xx}(\omega, k_x, z, z')$ defined by Eq. (15), we obtain

$$\chi_{xx,xx}(\omega, k_x, z, z') = \frac{g_{xxx}^u/\rho}{\omega_{TO}^2 - \omega^2 - i\omega\Gamma} \delta(z - z') - \frac{4\pi k_x^2}{\varepsilon(\omega)} \tilde{g}_{xxx}^E G(z, z') \quad (21)$$

To get the Raman cross-section, Eq. (14), we evaluate the integral

$$\int_{z_0}^{\infty} dz dz' e^{i(q_z z' - q_z^* z)} \text{Im} \chi_{ij,ij}(k, \omega, z, z').$$

We obtain for terms with $G(z, z')$

$$\text{Int} = \int_{z_0}^{\infty} dz f_-(z) \int_z^{\infty} dz' f_+(z') e^{i(q_z z' - q_z^* z)} + \int_{z_0}^{\infty} dz f_+(z) \int_{z_0}^z dz' f_-(z') e^{i(q_z z' - q_z^* z)},$$

and

$$\sum_{n,m=0}^{\infty} e^{i(q_z - q_z^*)nd + i(k_z - q_z)md} \int_{z_0}^{\infty} dz f_-(z) \int_z^{\infty} dz' f_+(z') e^{i(q_z z' - q_z^* z)} = \int_{z_0}^{z_0+d} dz f_-(z) \int_z^{z+d} dz' f_+(z') e^{i(q_z z' - q_z^* z)}. \quad (22)$$

The sum over the integer n could be extended to the infinity being equal to $\delta/2d$, if the thickness of the sample is larger compared with the skin-depth δ of the incident or scattered waves. The sum over m reduces to a not vanishing factor only under the Bragg condition, $k_z - q_z = 2\pi n/d$, which expresses the momentum conservation law in the exciting processes of the phonon-plasmon coupled modes. In the macroscopic limit, when the wave length of the exited mode is large compared to the period d , only the main Bragg maximum ($n = 0$) is observed for each of the coupled modes. In this case, we can expand in powers of $k_x d$ and $k_z d$ the matrix element in Eq. (22).

Omitting the common factors, we have the Raman intensity for the parallel geometry (a) in the form

$$Int_{xx}(\omega, k_x) = \text{Im} \left\{ \frac{g_{xxx}^2 / \rho}{\omega_{\text{TO}}^2 - \omega^2 - i\omega\Gamma} + \left(g_{xxx}^E + \frac{g_{xxx}^u Z / \rho}{\omega_{\text{TO}}^2 - \omega^2 - i\omega\Gamma} \right)^2 \frac{4\pi k_x^2}{(k_z^2 - q_z^2)\varepsilon(\omega)} \right\} \quad (23)$$

and for the crossed geometry (b)

$$Int_{xy}(\omega, k_x) = \text{Im} \left\{ \frac{g_{xyz}^2 / \rho}{\omega_{\text{TO}}^2 - \omega^2 - i\omega\Gamma} + \left(g_{xyz}^E + \frac{g_{xyz}^u Z / \rho}{\omega_{\text{TO}}^2 - \omega^2 - i\omega\Gamma} \right)^2 \frac{4\pi q_z^2}{(k_z^2 - q_z^2)\varepsilon(\omega)} \right\}. \quad (24)$$

The wave-vector k_z of the coupled phonon-plasmon mode is determined by Eq. (7). For example, if the incidence is normal to the sample surface and θ is the scattering angle, then $k_x = \omega^i \sin \theta / c$ and $q_z = (\sqrt{\varepsilon(\omega^i)} + \sqrt{\varepsilon(\omega^i) - \sin^2 \theta}) \omega^i / c$, where we take into account that $\omega^i \approx \omega^s$.

In Eqs. (23) – (24), we dropped slowly varying factors depending on the parameters of the incident and scattered radiations, for instance, their dielectric constants as well as the temperature factor $1/[1 + \exp(-\omega/T)]$. In numerical calculations, we used the relationship between the vertices g^u and g^E known from experiments and given by the Faust-Henry

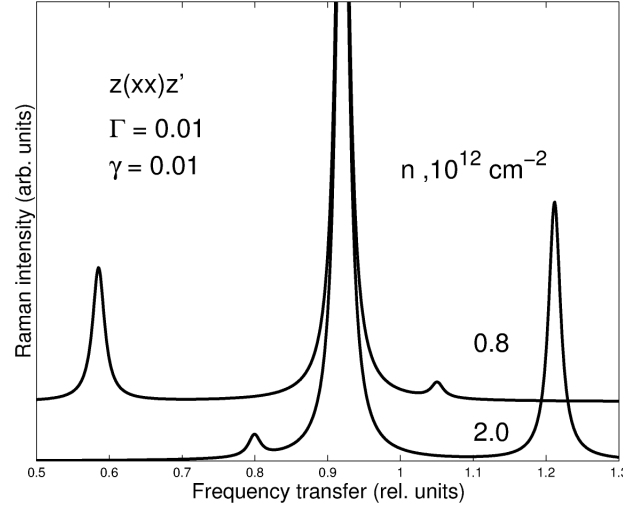


FIG. 13: Raman intensity vs. the frequency transfer (in units ω_{LO}) in the $z(xx)z'$ geometry for two values of the current concentration indicated at the curves in units of 10^{12} cm^{-2} ; z and z' are the propagation directions of the incident and scattered lights, respectively; the direction z' makes an angle of $\pi/4$ with the backscattering direction $-z$, (xx) are the corresponding polarizations. The values of the other parameters are the same as in Fig. 4.

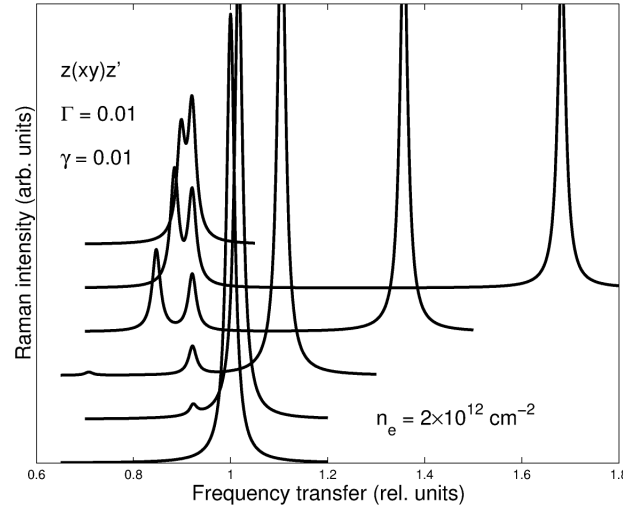


FIG. 14: Raman intensity vs. the frequency transfer (in units ω_{LO}) in the $z(xy)z'$ geometry for the angle of scattering varying from 0 (bottom) $\pi/2$ (top) with a step of $\pi/10$.

constant $C_{FH} = g^u Z / g^E \rho \omega_{TO}^2 = -0.5$. The results of calculations are shown in Figs. 13 and 14.

Note, that for the case of normal propagation of both incident and scattered radiation,

$k_x = 0$, the second term in Eq. (23) vanishes, and the Raman peak at parallel polarizations (geometry a) is situated at the frequency ω_{TO} . The others peaks in Fig. 13 correspond to the excitations of the coupled phonon-plasmons. But for the crossed polarizations (geometry b) and $k_x \rightarrow 0$, the dispersion equation (7) with $C \rightarrow 0$ gives $k_z = ik_x$. Then, using Eq. (24) and the relationship $\omega_{\text{LO}}^2 - \omega_{\text{TO}}^2 = 4\pi Z^2 / \epsilon_\infty \rho$ between the frequencies of the longitudinal and transverse phonons, we see that the peak appears only at ω_{LO} , since the terms with poles at ω_{TO} are cancelled. This peak corresponds to the zero of dielectric constant $\epsilon(\omega)$. At other scattering angles, a peak appears at ω_{TO} independently of the scattering angle. Two other peaks on each curves in Fig. 14 correspond to the excitation of the phonon-plasmons.

V. CONCLUSIONS

In this work, we investigated the infrared absorption and the Raman scattering on the coupled phonon-plasmon modes with the help of a simple model of superlattices formed by thin conducting layers separated with insulating layers. This model admits the dispersion relation of an analytical form. Our results for the reflectance and the Raman spectra show that the observed picture can be drastically modified by means of the carrier concentration, the superlattice period, and the frequency.

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