Acoustic plasmons in $Hg_{3-\delta}AsF_6$ in the long-wavelength limit

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In mercury chain compounds such as $Hg_{3-\delta}AsF_6$, the Hg atoms form two mutually perpendicular arrays of parallel chains (the *a* and *b* chains). Mohan has recently studied the random-phase-approximation coupled dynamic electronic response functions of a system in which the electrons are restricted to move in channels of such a coupled array. Besides the usual kind of bulk plasmonlike mode which propagates in the *a*-*b* plane, Mohan predicted a new acoustic plasmon in which the charge fluctuations in the two arrays are out of phase with each other. We give a simplified analysis in the long-wavelength limit (based on neglecting the effects of the chain lattice geometry) which allows a more transparent discussion of the properties of this acoustic plasmon.

Most quasi-one-dimensional conductors are composed of a three-dimensional array of parallel chains, the electrons being coupled through the intrachain and interchain Coulomb interaction. There is a considerable literature[1-4] on the full electronic response function of such a model of parallel chains, the interchain coupling being treated in the random-phase approximation (RPA). The plasmon frequency of the coupled array in the long-wavelength limit is given by

$$\omega^2(\mathbf{p})=\omega_P^2\frac{p_x^2}{p^2}+v_F^2p_x^2+\cdots . \tag{1}$$

Here p_x is the component of the plasmon wave vector $\mathbf{p}$ along the direction of the chains, v_F is the Fermi velocity, and ω_P is the bulk plasmon frequency. (Strictly speaking, as we discuss later, the coefficient of the second term is dependent[4] on the radius and spacing of the chains.)

Recently, Mohan[5] has given a model calculation of the dynamic electronic response function of *two* coupled mutually perpendicular arrays of chains, such as one finds in the mercury chain compound $Hg_{3-\delta}AsF_6$. It is known from x-ray and neutron scattering studies that the Hg atoms form two arrays (the so-called *a* and *b* chains) in which the Hg-Hg ion spacing is incommensurate with the host AsF_6 lattice.[6] Extensive theoretical[7] and experimental studies[6] have indicated that the important electronic states associated with the Hg chains can be approximated as one-dimensional (1D) plane-wave states with the energy $\hbar^2k^2/2m$, where k is the wave vector along the chain axis. If the hopping of electrons is ignored between different chains, one has a simple model for calculating the response function of the coupled chains in terms of the response function of a single chain, the interchain coupling being treated in the RPA. This model is an obvious generalization of earlier studies of a single array of parallel chains of finite radius.[2,4] In her thesis, Mohan derived the coupled equations of motion for the electronic response functions χ_{aa}, χ_{bb}, and χ_{ab} (where, for example, χ_{aa} involves the correlation function between the electronic density on two different chains in the *a* array) and predicted that, in addition to a plasmon analogous to (1), there was a new acoustic plasmon in which charge fluctuations in the *a* and *b* arrays are out of phase. However, the analysis given by Mohan is fairly complicated and as a result the underlying physics of the new acoustic plasmon is somewhat hidden. In the present paper, we present a new set of coupled equations of motion analogous to what one has in any 3D lattice and then discuss the simple long-wavelength limit in which we neglect all reciprocal-lattice vectors associated with the lattice of chains.

We first briefly review the model used by Mohan[5] (the details of which are also discussed in Ref. 4). The density fluctuation in a chain of the *a* array (along the *x* direction) is

$$\delta\rho^a(\mathbf{r},\omega)=\sum_{\mathbf{R}_a}\delta\rho_{\mathbf{R}_a}(x,\omega)w(\mathbf{r}_\perp-\mathbf{R}_a) , \tag{2}$$

where $\mathbf{r}_\perp$ is the component of $\mathbf{r}$ in the *yz* plane and $\mathbf{R}_a$ is the 2D-Bravais-lattice vector of the *a*-chain lattice. The electrons in a given chain are assumed to be in their lowest transverse state. In the Gaussian approximation[2,4] this chain leads to the form factor $w(\mathbf{r}_\perp)=(\pi r_0^2)^{-1}\exp(-r_\perp^2/r_0^2)$, where r_0 is the effective radius of the cylindrical chain potential.

Within linear response, we have

$$\delta\rho^n(\mathbf{r},\omega)=\sum_{m=a,b}\int d\mathbf{r}'\chi_{nm}(\mathbf{r},\mathbf{r}';\omega)V_0^m(\mathbf{r}',\omega) , \tag{3}$$

where $V_0^m(\mathbf{r}',\omega)$ describes the external potential acting on electrons in the m array and

$$\chi_{nm}(\mathbf{r},\mathbf{r}',\omega) = -i\int_0^\infty dt\, e^{i(\omega+i0^+)t}\langle[\delta\hat{\rho}^n(\mathbf{r},t),\delta\hat{\rho}^m(\mathbf{r}',0)]\rangle . \tag{4}$$

Within the RPA, we have

$$\delta\rho^n(\mathbf{r},\omega)=\int d\mathbf{r}'\chi_{nn}^0(\mathbf{r},\mathbf{r}',\omega)V_{\rm tot}^n(\mathbf{r}',\omega) , \tag{5}$$

where the self-consistent field acting on the electrons in a chain of the nth array is[5]

$$V^n_{\rm tot}(\mathbf{r}',\omega) = V^n_0(\mathbf{r}',\omega)+\sum_{m=a,b}\int d\mathbf{r}''\, v_{n,m}(\mathbf{r}'-\mathbf{r}'')\delta\rho^m(\mathbf{r}'',\omega)\ . \quad (6)$$

Here $v_{n,m}$ is the (Coulomb) interaction between electrons on chains in the n and m arrays. In the above treatment, the intrachain Coulomb interactions are also being treated in the RPA (although this is not necessary).

We make crucial use of the properties of the electron response functions $\chi^0_{m,n}$ in the absence of all Coulomb interactions. Only electrons in a *given* chain are correlated and thus

$$\begin{aligned}
&\chi^0_{ab}(\mathbf{r},\mathbf{r}';\omega)=0\ ,\\
&\chi^0_{aa}(\mathbf{r},\mathbf{r}';\omega)=\sum_{\mathbf{R}_a} w(\mathbf{r}_\perp-\mathbf{R}_a)w(\mathbf{r}'_\perp-\mathbf{R}_a)\chi^0_{\mathbf{R}_a\mathbf{R}_a}(x-x';\omega)\ ,\\
&\chi^0_{bb}(\mathbf{r},\mathbf{r}';\omega)=\sum_{\mathbf{R}_b} w(\mathbf{r}_\perp-\mathbf{R}_b)w(\mathbf{r}'_\perp-\mathbf{R}_b)\chi^0_{\mathbf{R}_b\mathbf{R}_b}(y-y';\omega)\ .
\end{aligned} \quad (7)$$

Here $\chi^0_{\mathbf{R}_a\mathbf{R}_a}(x-x')$ is the same for all a chains and we shall simply use χ^0 to denote the noninteracting response function for a strictly 1D electron gas. In (7), we emphasize that $\mathbf{r}_\perp$ is a vector in the plane perpendicular to the chain in question.

Substituting (6) into (5) gives a closed set of equations which can be most conveniently solved by using Fourier transforms (we leave the frequency dependence on ω implicit):

$$\chi_{mn}(\mathbf{p},-\mathbf{q})\equiv\int d\mathbf{r}\, e^{i\mathbf{p}\cdot\mathbf{r}}\int d\mathbf{r}' e^{-i\mathbf{q}\cdot\mathbf{r}'}\chi_{mn}(\mathbf{r},\mathbf{r}')\ . \quad (8)$$

We note that the localization of the electrons into chains comes in automatically through the form factors $w(\mathbf{r}_\perp-\mathbf{R}_{a,b})$ in (7). In contrast with the analysis of Ref. 5, we proceed with a straightforward calculation identical to the usual one for the RPA electronic response function for electrons moving in a 3D periodic arrays of ions. We obtain

$$\delta\rho^n(\mathbf{p})=\sum_{\mathbf{q},m}\chi_{nm}(\mathbf{p},-\mathbf{q})V^m_0(\mathbf{q}) \quad (9)$$

and

$$\begin{aligned}
\delta\rho^a(\mathbf{p})=&\sum_{\mathbf{q}}\chi^0_{aa}(\mathbf{p},-\mathbf{q})\sum_{\mathbf{G}_a}\delta_{\mathbf{q},\mathbf{p}+\mathbf{G}_a}V^a_0(\mathbf{q})+\chi^0(p_x)\beta(\mathbf{p}_\perp)\sum_{\substack{\mathbf{q},\mathbf{G}_a,\\ m}}[v_{aa}(\mathbf{p}+\mathbf{G}_a)\beta(\mathbf{p}_\perp+\mathbf{G}_a)\chi_{am}(\mathbf{p}+\mathbf{G}_a,-\mathbf{q})V^m_0(\mathbf{q})\\
&+v_{ab}(\mathbf{p}+\mathbf{G}_a)\beta(\mathbf{p}_\perp+\mathbf{G}_a)\chi_{bm}(\mathbf{p}+\mathbf{G}_a,-\mathbf{q})V^m_0(\mathbf{q})]\ .
\end{aligned} \quad (10)$$

Here $\mathbf{p}_\perp$ is the component of $\mathbf{p}$ in the yz plane and $\mathbf{G}_a$ is a 2D reciprocal-lattice vector of the $\mathbf{R}_a$ lattice. $\beta(\mathbf{p}_\perp)$ is the Fourier transform of $w(\mathbf{r}_\perp)$ and, in our Gaussian approximation, this is given by $\beta(\mathbf{p}_\perp)=\exp(-p_\perp^2 r_0^2/4)$. Key results used in deriving (10) are that[2–4]

$$\chi^0_{aa}(\mathbf{p},-\mathbf{q})=0 \quad \text{unless } \mathbf{q}=\mathbf{p}+\mathbf{G}_a \quad (11)$$

and

$$\chi^0_{aa}(\mathbf{p},-\mathbf{p}-\mathbf{G}_a)=\beta(\mathbf{p}_\perp)\chi^0(p_x)\beta(\mathbf{p}_\perp+\mathbf{G}_a)\ . \quad (12)$$

It is the condition (11) which is the source of the restricted sums over $\mathbf{G}_a$ in (10).

A result identical to (10) can be derived for $\delta\rho^b(\mathbf{p},\omega)$, with a and b labels interchanged as well as $p_x\to p_y$. In addition, $\mathbf{p}_\perp$ is now the component in the xz plane and expressions analogous to (11) and (12) also hold for χ^0_{bb}.

Combining (9) and (10), and making use of the fact that (A_c is the area of the unit cell of the chain lattice)

$$v_{aa}(\mathbf{q})=v_{bb}(\mathbf{q})=v_{ab}(\mathbf{q})=\frac{4\pi e^2}{A_c q^2}\equiv v(\mathbf{q})\ , \quad (13)$$

we obtain

$$\chi_{an}(\mathbf{p},-\mathbf{q})=\chi^0_{aa}(\mathbf{p},-\mathbf{q})\sum_{\mathbf{G}_a}\delta_{\mathbf{q},\mathbf{p}+\mathbf{G}_a}\delta_{an}+\chi^0(p_x)\sum_{\mathbf{G}_a}\beta(\mathbf{p}_\perp)v(\mathbf{p}+\mathbf{G}_a)\beta(\mathbf{p}_\perp+\mathbf{G}_a)[\chi_{an}(\mathbf{p}+\mathbf{G}_a,-\mathbf{q})+\chi_{bn}(\mathbf{p}+\mathbf{G}_a,-\mathbf{q})] \quad (14)$$

and a similar equation for $\chi_{bn}(\mathbf{p},-\mathbf{q})$, with a and b labels interchanged and $p_x\to p_y$. We remark that if the *inter*array coupling v_{ab} is omitted in (10), (14) can be solved to give the results for a single array[2,4]

$$\chi_{aa}(\mathbf{p},-\mathbf{p}-\mathbf{G}_a)=\frac{\chi^0_{aa}(\mathbf{p},-\mathbf{p}-\mathbf{G}_a)}{1-u(\mathbf{p})\chi^0(p_x)}\ , \quad (15)$$

where the form-factored Coulomb potential is

$$u(\mathbf{p})\equiv\sum_{\mathbf{G}_a}|\beta(\mathbf{p}_\perp+\mathbf{G}_a)|^2 v(\mathbf{p}+\mathbf{G}_a)\ . \quad (16)$$

By a direct consideration of the symmetry of the coupled arrays, one may show that

$$\begin{aligned}
&\chi_{\mathbf{R}_a+\bar{Y}_a,\mathbf{R}_b}(x^a,y^b+\bar{Y}_a)=\chi_{\mathbf{R}_a,\mathbf{R}_b}(x^a,y^b)\ ,\\
&\chi_{\mathbf{R}_a,\mathbf{R}_b+\bar{X}_b}(x^a+\bar{X}_b,y^b)=\chi_{\mathbf{R}_a,\mathbf{R}_b}(x^a,y^b)\ ,\\
&\chi_{\mathbf{R}_a+\bar{Z}_a,\mathbf{R}_b+\bar{Z}_a}(x^a,y^b)=\chi_{\mathbf{R}_a,\mathbf{R}_b}(x^a,y^b)\ ,
\end{aligned} \quad (17)$$

and

$$\begin{aligned}
&\chi_{\mathbf{R}_a,\mathbf{R}'_a}(x,x')=\chi_{\mathbf{R}_a-\mathbf{R}'_a}(x-x')\ ,\\
&\chi_{\mathbf{R}_b,\mathbf{R}'_b}(y,y')=\chi_{\mathbf{R}_b-\mathbf{R}'_b}(y-y')\ .
\end{aligned} \quad (18)$$

Here $\bar{X}_b$, $\bar{Y}_a$, and $\bar{Z}_a$ are lattice translation vectors of the appropriate array along the x, y, and z axis, respectively.

Fourier transforming (17) and (18) leads to

$$\chi_{aa}(\mathbf{p},-\mathbf{q})=0 \quad \text{unless } \mathbf{q}=\mathbf{p}+\mathbf{G}_a \ ,$$
$$\chi_{bb}(\mathbf{p},-\mathbf{q})=0 \quad \text{unless } \mathbf{q}=\mathbf{p}+\mathbf{G}_b \ , \tag{19}$$
$$\chi_{ab}(\mathbf{p},-\mathbf{q})=0 \quad \text{unless } \mathbf{q}=\mathbf{p}+\mathbf{G}_a+\mathbf{G}_b \ .$$

As expected, $\chi_{ab}(\mathbf{p},-\mathbf{q})$ is completely symmetric in the dependence on the properties of the two arrays. In deriving (19), we made use of the fact that both $\mathbf{G}_a$ and $\mathbf{G}_b$ vectors share the same z components and thus the z component of $\mathbf{G}_a+\mathbf{G}_b$ is the z component of some vector $\mathbf{G}'_a$. One may verify that the RPA equations of motion such as in (14) are consistent with (19).

The coupled equations given by (14) and their equivalent for $\chi_{b,n}$ are very complicated because of the effects of the periodic arrays of chains (this is the origin of the sums over reciprocal-lattice vectors $\mathbf{G}_a$ and $\mathbf{G}_b$). In Mohan's work,[5] a general solution of a similar set of coupled equations is discussed in terms of form-factored potentials such as (16). In contrast, we discuss the solution of (14) starting right from the beginning with the neglect of local-field corrections, that is, we only keep $\mathbf{G}_a=0$ and $\mathbf{G}_b=0$ in the summations. This corresponds to the limit $pd \ll 1$, where d is the lattice spacing and, for consistency, we set $\beta(\mathbf{p}_\perp)=1$ since $r_0 \ll d$. In this long-wavelength limit, the coupled equations in (14) reduce to

$$\chi_{an}(\mathbf{p},-\mathbf{p})=\chi^0(p_x)\delta_{an} + \chi^0(p_x)v(\mathbf{p})[\chi_{a,n}(\mathbf{p},-\mathbf{p})+\chi_{b,n}(\mathbf{p},-\mathbf{p})] \ ,$$
$$\chi_{bn}(\mathbf{p},-\mathbf{p})=\chi^0(p_y)\delta_{b,n} + \chi^0(p_y)v(\mathbf{p})[\chi_{b,n}(\mathbf{p},-\mathbf{p})+\chi_{a,n}(\mathbf{p},-\mathbf{p})] \ . \tag{20}$$

Solving these, we see that the poles of all the response functions are given by the zeros of the dielectric function

$$\lim_{\mathbf{p}\to 0}\epsilon(\mathbf{p},\omega)=1-v(\mathbf{p})[\chi^0(p_x,\omega)+\chi^0(p_y,\omega)] \ . \tag{21}$$

This elegantly simple result is the basis of the rest of our analysis. It could have been written down almost without calculation since it just says that the usual 3D Lindhard function is "contracted down" by the restriction of motion to the x and y directions.

The response function for a noninteracting degenerate 1D Fermi gas in the long-wavelength limit (q much less than the Fermi momentum) is given by[2-4,8]

$$\chi^0(q,\omega)\simeq N(\varepsilon_F)\frac{v_F^2q^2}{\omega^2-v_F^2q^2} \ . \tag{22}$$

Here the 1D electronic density of states is $N(\varepsilon_F)=2/\pi\hbar v_F$ and the Fermi velocity is $v_F=N_e\hbar\pi/2Lm$, where N_e/L is the number of electrons per unit chain length. Using (22) in (21), we obtain

$$\epsilon(\mathbf{p},\omega)=1-\frac{\omega_P^2}{p^2}\left[\frac{p_x^2}{\omega^2-v_F^2p_x^2}+\frac{p_y^2}{\omega^2-v_F^2p_y^2}\right] , \tag{23}$$

where $\omega_P^2=4\pi ne^2/m$ with $n\equiv N_e/A_cL$. For a given value of $\mathbf{p}$, we see that the highly degenerate free-electron chain modes $\omega_a^0(\mathbf{p})=v_Fp_x$ and $\omega_b^0(\mathbf{p})=v_Fp_y$ are coupled and renormalized, the new modes being given by the zeros of $\varepsilon(\mathbf{p},\omega)$. The $\omega_{a,b}^0$ modes play the role of particle-hole excitations in the well-known analysis of $\varepsilon(\mathbf{p},\omega)$ in a 3D electron gas. The significance of the two renormalized modes $\omega_\pm(\mathbf{p})$ can be most easily understood from the graphical plot of $1-\varepsilon(\mathbf{p},\omega)$ versus ω shown in Fig. 1. Assuming that p_x and p_y are both finite and that $p_x<p_y$, one sees that

$$v_Fp_x<\omega_-(\mathbf{p})<v_Fp_y\ll\omega_+(\mathbf{p}) \ . \tag{24}$$

A direct analysis of (23) shows that *only* the $\omega_+(p)$ mode exists if (a) either p_x *or* $p_y=0$ or (b) $p_x=p_y$. This makes sense since in both cases, there is only a single frequency associated with the noninteracting single chains.

In the long-wavelength limit, the zeros of $\varepsilon(\mathbf{p},\omega)$ are easily obtained and we find (for $p_x\neq p_y$)

$$\omega_+^2(\mathbf{p})\simeq\omega_P^2\frac{p_x^2+p_y^2}{p^2}+v_F^2(p_x^2+p_y^2)-2v_F^2\frac{p_x^2p_y^2}{p_x^2+p_y^2}$$
$$=\omega_P^2\sin^2\theta+v_F^2p^2\sin^2\theta[1-\tfrac{1}{2}\sin^2(2\phi)] \ , \tag{25}$$

$$\omega_-^2(\mathbf{p})\simeq 2v_F^2\frac{p_x^2p_y^2}{p_x^2+p_y^2}$$
$$=\tfrac{1}{2}v_F^2p^2\sin^2\theta\sin^2(2\phi) \ , \tag{26}$$

where $\mathbf{p}=(p,\theta,\phi)$ gives the wave vector is spherical coordinates. If we ignore the effect of the chain lattice geometry and the finite radius of the chains in the dispersion relations given in Ref. 5, they reduce to (25) and (26) in the long-wavelength limit. The $\sin^2\theta$ factor emphasizes that both modes only involve the component of $\mathbf{p}$ in the a-b (or x-y) plane. The absence of the $\omega_-(\mathbf{p})$ mode when either p_x or p_y is zero indicates that this

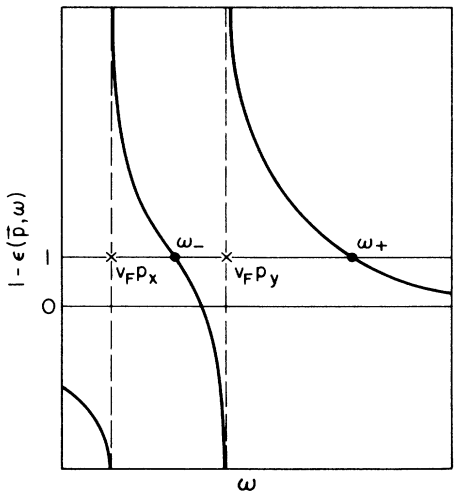

FIG. 1. A schematic plot of the electronic dielectric function of coupled arrays [as given by Eq. (23)] vs frequency. This shows how the charge fluctuations of two 1D noninteracting Fermi gases (v_Fp_x,v_Fp_y) are coupled and renormalized to become the interarray plasmons $\omega_\pm(\mathbf{p})$.

mode is intrinsically an interarray plasmon mode. In the special case $p_x = p_y$, one can show that only the ω_+ mode exists, with a dispersion relation given by (25) with $\phi = 45°$ (the absence of the acoustic plasmon for $p_x \simeq p_y$ was not noted in Ref. 5).

Using the equations in (20), one can easily verify that the ratio of charge fluctuations in the two arrays is

$$\frac{\delta\rho^a(\mathbf{p},\omega)}{\delta\rho^b(\mathbf{p},\omega)} = \frac{\chi^0(p_x,\omega)}{\chi^0(p_y,\omega)} = \frac{p_x^2}{p_y^2}\left[\frac{\omega^2 - v_F^2 p_y^2}{\omega^2 - v_F^2 p_x^2}\right] . \tag{27}$$

Inserting (25) in (27), one finds that the bulklike plasmon ω_+ involves a relative oscillation of the two arrays given by

$$\frac{\delta\rho^a(\mathbf{p},\omega_+)}{\delta\rho^b(\mathbf{p},\omega_+)} \simeq \frac{p_x^2}{p_y^2} . \tag{28}$$

It is only in the special case $p_x = p_y$ that the two arrays oscillate perfectly in phase. In contrast, using (26) in (27) gives $\delta\rho^a(\mathbf{p},\omega_-) = -\delta\rho^b(\mathbf{p},\omega_-)$. That is, the long-wavelength acoustic plasmon ω_- always involves charge fluctuations in the two arrays which are completely out of phase. This means that in this limit, the ω_- mode will have vanishingly small weight in the dynamic structure factor since no *net* density fluctuation is involved. A more detailed analysis shows that the acoustic plasmon does involve a net density fluctuation at larger wave vectors.

The preceding analysis of the interarray plasmons $\omega_\pm(\mathbf{p})$ has been based on Eq. (20). We feel this continuum approximation brings out the essential physics. However, Mohan's analysis shows that the coefficients of the p^2 terms in (25) and (26) do have corrections which depend on the form factors $\beta(\mathbf{p}_\perp)$ as well as the lattice structure of the chains. The kind of modification can be illustrated in the much simpler case of a *single* array.[2,4] As described by (15), the dependence on r_0 and d is completely contained in the form-factored Coulomb potential in (16). In the limit $d \gg r_0$, $pd \ll 1$, one finds[9]

$$u(\mathbf{p}) = e^2\left[\frac{4\pi}{A_c p^2} + \ln\left[\frac{a^2}{2r_0^2}\right] + \gamma - 1 + O(p_x^2)\right] , \tag{29}$$

where $A_c \equiv \pi a^2 = d^2\sqrt{3}/2$ and $\gamma \simeq 0.577$ is Euler's constant. The resulting intra-array plasmon dispersion relation is given by

$$\omega^2(\mathbf{p}) = \omega_P^2\frac{p_x^2}{p^2} + v_F^2 p_x^2\left\{1 + e^2 N(\varepsilon_F)\left[\ln\left[\frac{a^2}{2r_0^2}\right] + \gamma - 1\right]\right\} \tag{30}$$

instead of (1). For $Hg_{3-\delta}AsF_6$, $e^2N(\varepsilon_F)$ is of order unity. Similar modifications to (25) and (26) are expected, of the kind discussed in Ref. 5.

To the extent that we work in the long-wavelength limit described by Eq. (23), there is no Landau damping of the acoustic plasmon $\omega_-(\mathbf{p})$. Formally this can be seen from the fact that the only imaginary contributions to (23) occur at the free-particle chain modes at $v_F p_x$ and $v_F p_y$. As is clear from Fig. 1, neither is degenerate with the acoustic plasmon. In a more realistic model, one expects that the ω_- mode will be strongly damped in the region around $\phi = 45°$ ($p_x \simeq p_y$).

Experimental observation of the new acoustic plasmon ω_- would confirm the basic correctness of the simple model for the response functions of the coupled chains introduced by Mohan.[5] We might note that in the somewhat analogous theoretical problem of coupled-layer plasmons[10] in semiconductor superlattices, Raman scattering has been used successfully.[11] For $p_\parallel \sim 10^5$ cm^{-1}, the acoustic plasmon in (26) has an energy of the order of 10 meV. However, we recall that the acoustic plasmon ω_- is only weakly coupled into the density fluctuation spectrum in the long-wavelength limit. It is not clear what sort of experimental probe is appropriate.

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