Plasmons in aperiodic structures

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We study and compare plasmon spectra in semiconductor superlattices with periodic, quasiperiodic (e.g., Fibonacci sequence), and random spacings. Novel mode structures are found in the quasiperiodic cases, particularly for low values of level broadening that arises from inherent disorder effects. We discuss critically the observability of this novel mode structure via light scattering studies in experimentally realizable superlattices. In addition, our results suggest that the finite Fibonacci-sequence superlattice may be considered as a partially ordered-layer lattice with no long-range positional order, but with strong short-range order, since our calculated plasmon spectra exhibit very little difference between the finite Fibonacci sequence and the periodic *abaab* superlattice.

Plasmons in semiconductor superlattices have attracted great theoretical[1-5] and experimental[6-9] attention recently.[10] Part of the reason[10] for this interest is the possibility of studying collective modes in reduced dimensionality and comparing experimentally observed plasmon dispersion relation (and spectral weight) with many-body theoretical linear-response calculations.[1,2] There is also great interest in the prediction (and, hopefully, eventual observation) of novel collective modes in artificially structured superlattices which are not found in bulk systems. Thus, discrete plasmons,[2,11] surface[3] and edge[4] plasmons, and localized and critical plasmon modes[5] have been predicted in various superlattice structures. Discrete plasmons in a finite-layered structure have recently been observed by two different groups[9] in Raman scattering measurements. It is expected that with the advance in materials preparation and fabrication techniques, many more such studies will appear.

In our earlier paper,[5] we pointed out that artificially structured random and incommensurate superlattices are ideal candidates for studying localization effects in one-dimensional systems via Raman scattering studies of their collective mode spectra. We showed[5] that Raman scattering directly measures the local plasmon ("one-particle") density of states in **k** space, and that the plasmon problem in a random or an incommensurate superlattice is formally equivalent to the Anderson or the Aubry model of localization, respectively. We proposed specific experiments to be carried out on artificial periodic structures where the layer electron density varies in a random or quasiperiodic fashion, and predicted that distinct localization effects will show up in such light scattering experiments. Experimentally, an easier system to fabricate is a structure where the layers themselves are placed in random or quasiperiodic positions along the superlattice growth axis (taken to be the z axis). In fact, a quasiperiodic GaAs-AlAs superlattice in which the layers are placed in a Fibonacci sequence has recently been constructed by Merlin *et al.*[12] In this paper, we study the collective modes in such aperiodic structures and discuss to what extent any of the novel and rich physics predicted[13] for quasiperiodic one-dimensional systems (e.g., Cantor set spectrum, existence of extended, localized, and singular continuous states) will be manifested in the *actual experimental system.* Since the basic mapping of the plasmon problem into the equivalent Anderson Hamiltonian has already been explained in our earlier paper,[5] we shall concentrate in this paper on the question of physical resolution limitations arising from the damping (and the consequent broadening) of the plasmons, which imposes severe restrictions on the observability of novel "quasiperiodic physics" in such Fibonacci-sequence superlattices. Our finding is that one must have low values of broadening to see a distinct difference between the plasmon spectra in a Fibonacci superlattice and in a periodic superlattice via direct frequency-scan Raman measurement. (Raman spectrum obtained in **k** space is much more useful for this purpose.[5]) Although systematic studies of plasmon modes in a wide class of superlattices including random, periodic, and quasiperiodic structures of different sorts will clearly show signatures of novel quasiperiodic physics (as we discuss below), a quantitative study of the Cantor set spectrum and scaling behavior[13] seems unlikely due to finite-size effects and the disorder-induced resolution limitations inherent in semiconductor superlattices.

In many ways our current study of plasmons in aperiodic structures is complementary to our earlier work,[5] where the case of a continuous incommensurate or random modulation of electron density in equally spaced layers was considered. In the present calculation we keep the electron density constant in all layers, whereas the layers themselves are placed in some aperiodic fashion. We should emphasize that randomness or quasiperiodicity enters into the formalism differently in these two situations: in the present case aperiodicity enters through an exponential since the layer separation only affects the interlayer Coulomb interaction, whereas in our earlier work[5] randomness or incommensuration entered through the electron density which appears algebraically in the electronic polarizability function of the layered system.[14] The motivation for the current analysis stems from (i) the actual fabrication of a Fibonacci superlattice as reported in Ref. 12 and (ii) the fact that aperiodic multilayer structures based on random or quasiperiodic positioning of individual layers (with constant electron density) are perhaps easier to make than structures with the artificially predesigned electron-density modulation which we considered in Ref. 5.

Since extensive numerical studies on quasiperiodic one-dimensional systems exist in the literature,[13] we concentrate in this paper on the experimental observability in actual semiconductor superlattices, and consider directly the experimentally relevant (and directly measured) Raman scattering intensity (which is also the local plasmon density of states in $\mathbf{k}$ space[5]). The Raman intensity is related to the dynamical structure factor of the layered electron gas, and is proportional to

$$I(q,k,\omega)=\sum_{j,j'}\mathrm{Im}[\Pi^{-1}-V]^{-1}{}_{jj'}\exp[ik(z_j-z_{j'})]\ , \qquad (1)$$

where Π is the dynamic electron polarizability and $V_{jj'}=(2\pi e^2/\epsilon q)\exp(-q\,|z_j-z_{j'}|)$ is the Coulomb interaction between electrons in layers j and j' (with ϵ as the average background dielectric constant). We take z_j to be the position of the jth layer, and (q,k,ω) is the wavenumber-frequency difference of incident and outgoing photons with q as the conserved wave vector in the x-y plane and k as the charge in the photon's wave vector in the z direction. We assume all the layers have the same electron density, $n_j=N_0$, whereas their positions z_j could be random, quasiperiodic, or periodic. (In our earlier analysis,[5] we took z_j to be periodic and n_j as aperiodic.[14])

Given a choice for z_j, one can readily calculate the Raman intensity, $I(q,k,\omega)$ of Eq. (1). We take the total number of layers to be 25 in this paper, except for the Fibonacci system which has 34 layers. (This choice is motivated by the actual systems of Refs. 6—9.) One can, of course, study much larger systems numerically, and we have done so. However, these studies are of no experimental relevance since light can penetrate only a finite number of layers (usually of the order of ten layers) due to extinction effect. For the sake of comparison, we have also shown the infinite periodic superlattice case in Fig. 1. In Figs. 2—5, we show our calculated Raman intensity for different models of z_j and for three different values of the broadening (Γ) in each case. In an ideal system, the broadening arises from finite-size effects which one can study systematically by using a finite-size scaling analysis. In systems of our interest,[6–9,12] the main contribution to broadening is the inherent disorder effect always present in semiconductor superlattices. Thus the spectral peaks of Eq. (1), instead of being δ-function-like, broaden out into bands and the broadening enters the theory via the dynamic polarizability, Π in Eq. (1), which we calculate in a Mermin-type[15] generalization of the two-dimensional Stern's formula.[16] The broadening parameter Γ sets the limit for the ultimate resolution with which the discrete spectrum of Eq. (1) can be resolved. Thus Γ is the critical parameter determining the resolution of the Cantor-set spectrum. We discuss the origin of Γ later in the paper. In Fig. 1 we show our calculated Raman intensity for the periodic system, $z_j=jd$ where d is the period, for both the infinite system[1] and the finite 25-layer system. In Fig. 2 we show our calculated Raman intensity for the Fibonacci-sequence superlattice[12] which has fundamental periods a and b $(=d)$ with the ratio $a/b=(5^{1/2}+1)/2=\tau$, the golden mean, and the aperiodic sequence $abaab\ldots$. Also shown as an inset in Fig. 2(c) is the plasmon-dispersion relation for the Fibonacci system. In Fig. 3 we show our results for the incommensurate 25-

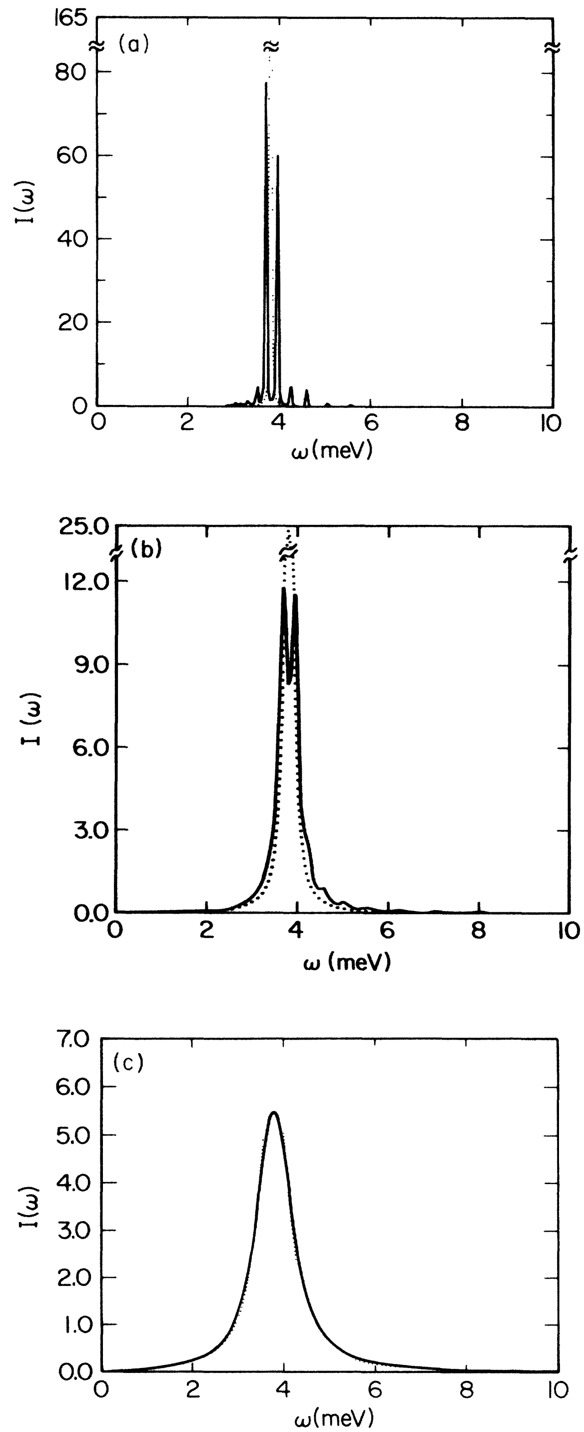

FIG. 1. The Raman intensity $I(q,k,\omega)$ as a function of frequency ω (for fixed $qd=0.43$ and $kd=4.94$) for the finite 25-layer (solid line) and the infinite (dotted line) periodic systems for three values of broadening parameter Γ: (a) 0.02, (b) 0.2, and (c) 0.7 meV.

layer structure with $z_j = d\{1+0.25[\cos(j)+\cos(\tau j)]\}$. In Fig. 4 we show our results for the random superlattice with $z_j = d(1+R_j)$, where R_j is a random number between 0.5 and -0.5. We show results for three different values of Γ: (a) 0.02, (b) 0.2, and (c) 0.7 meV. (These values are chosen to range from far too optimistic through realistic to rather pessimistic situations.) The intensity in all figures is depicted on the same absolute units so that a comparison between them is meaningful. The length scale (or the average period) of the superlattices is chosen to be $d = 890$ Å, and we take $N_0 = 7.3\times10^{11}$ cm^{-1} with $qd = 0.43$ and $kd = 4.94$. These parameters are fairly typical for superlattice plasmon experiments and are taken from Ref. 6. (Typically qd varies between 0.1 and 1.5 whereas kd could vary between 3 and 7.) Qualitative features of our results are independent of any specific

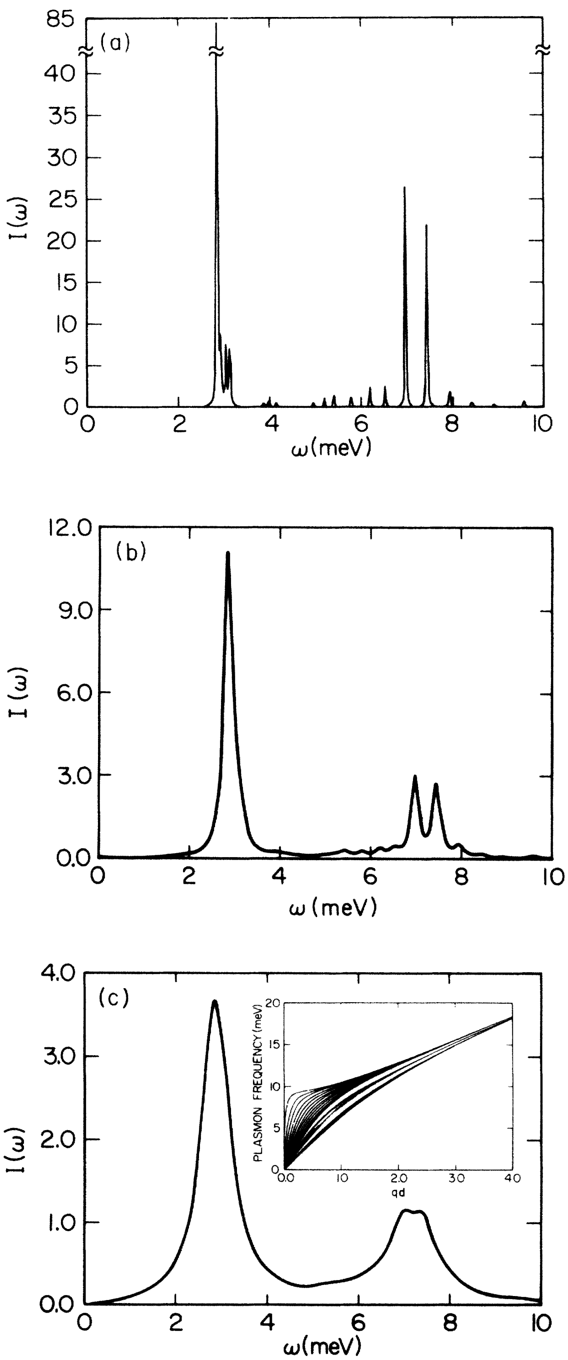

FIG. 2. The Raman intensity $I(q,k,\omega)$ as a function of frequency ω for the 34-layer Fibonacci-sequence superlattice for the same values of the parameters (q, k, and Γ) as in Fig. 1. Also shown as an inset in (c) is the plasmon dispersion relation.

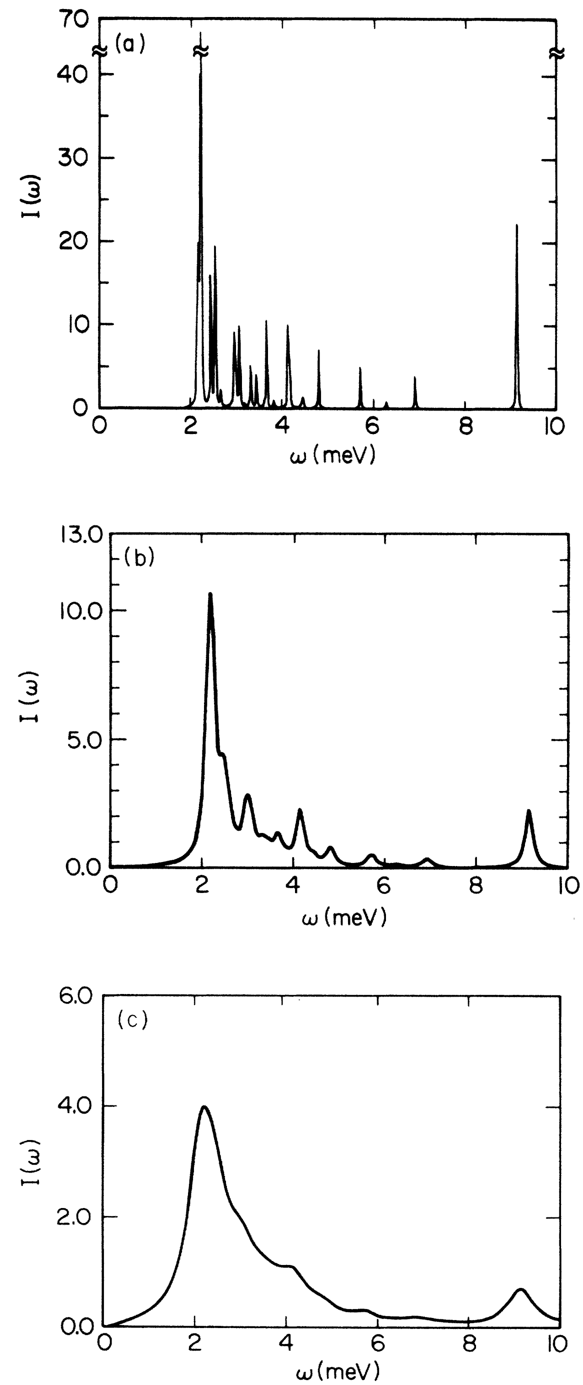

FIG. 3. The Raman intensity $I(q,k,\omega)$ as a function of frequency ω for the 25-layer quasiperiodic cosine superlattice system with the same values of parameters as in Figs. 1 and 2.

choice of these parameters, provided the parameters are in the experimentally accessible range. It should be mentioned here that our numerical result for the periodic system (Fig. 1) with $\Gamma=0.7$ meV is in good agreement (within 5%) with the measured[6] Raman scattering spectrum.

From Figs. 1–4, we conclude that even though the plasmon spectra in the *pure* case ($\Gamma\cong 0$) are fundamentally different in the periodic and the aperiodic structures, the presence of a finite damping effect substantially reduces the resolution so that the Cantor-set spectrum of plasmons in Fibonacci structures is, in general, not observable in real systems. Theoretically, the plasmon modes in the idealized periodic case are extended, the random situation is localized (the localization length, howev-

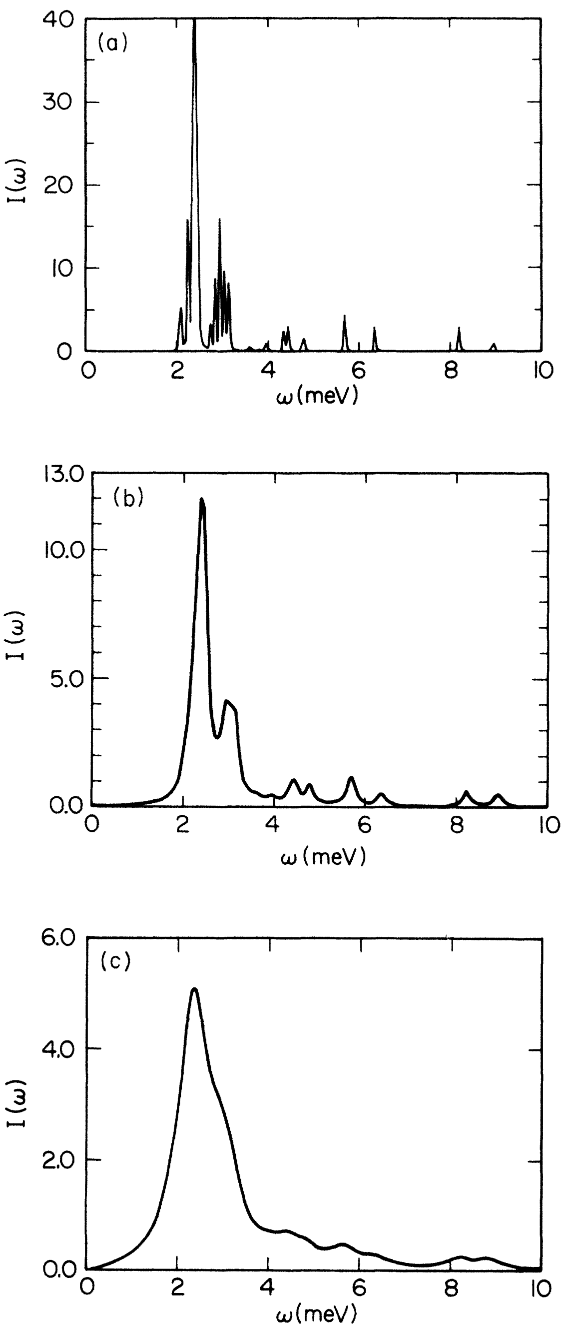

FIG. 4. The Raman intensity $I(q,k,\omega)$ as a function of frequency ω for the random 25-layer superlattice system with the same values of parameters as in Figs. 1, 2, and 3.

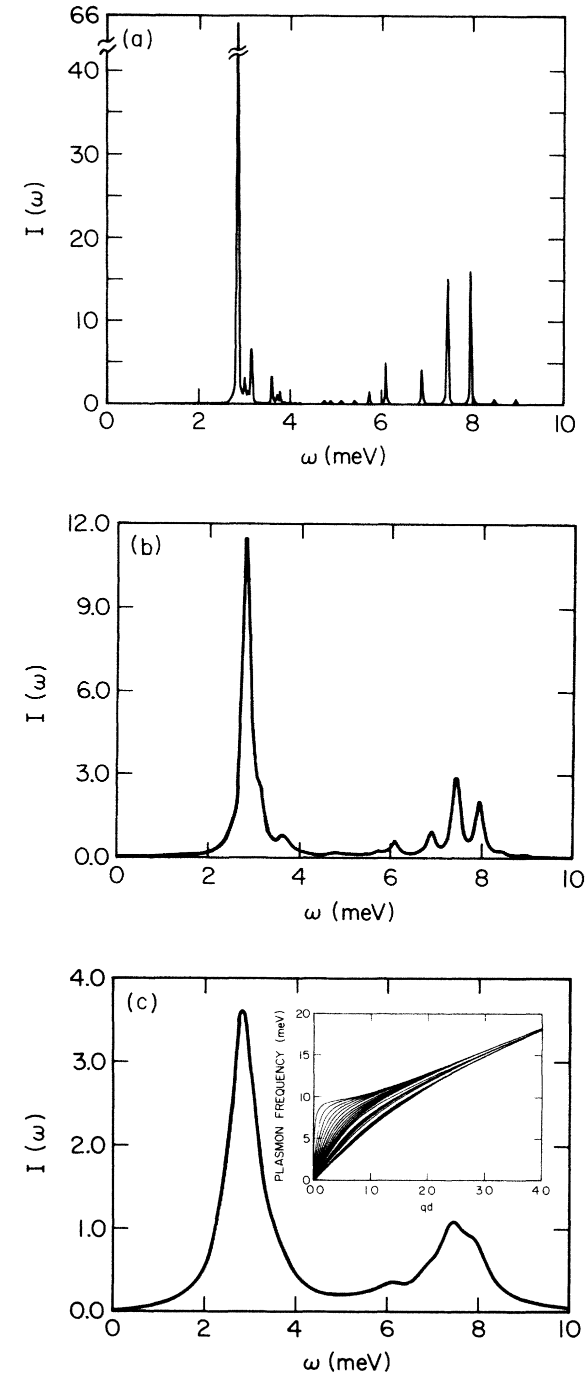

FIG. 5. The Raman intensity $I(q,k,\omega)$ as a function of frequency ω for the periodic *abaab* superlattice (see text) with the same values of parameters as in Figs. 1–4. The number of layers chosen for this case is 34, instead of 35, to compare the results with those of the Fibonacci-sequence superlattice (Fig. 2). We have found very little difference in spectra between the 34-layer and 35-layer *abaab* superlattices.

er, depends on which eigenstate one is looking at, and could be larger than the system size), whereas the quasiperiodic case is intermediate. In the ideal infinite situation, all the modes of the Fibonacci sequence are singular continuous (neither localized nor extended), whereas our model aperiodic system (Fig. 3) allows for point (localized), continuous (extended), and singular continuous (critical) solutions depending on the specific eigenmode one is considering. The experimental difference, on the other hand, is crucially dependent on the magnitude of Γ and can only be resolved uniquely if one obtains the spectrum in $\mathbf{k}$ space as we have emphasized earlier.[5] We point out the fact that the finite-damping effect and the finite number of layers participating in the light scattering experiments inherently limit the resolution of the plasmon spectra. However, as is obvious from our results, in relatively clean systems (small Γ) one can observe fundamental differences in the plasmon spectra between the periodic system (Fig. 1) and the aperiodic systems (Figs. 2–4) on the one hand, and, between the random (Fig. 4) and quasiperiodic situations (Figs. 2 and 3) on the other hand. This is particularly true at low values of level broadening.

Before concluding, we would like to provide some understanding of the Fibonacci spectra shown in Fig. 2. Our 34-layer Fibonacci sequence looks like

$$|\,abaab\,|\,abaab\,|\,aab\,|\,abaab\,|\,abaab\,|\,aab\,|\,abaab\,|\,aab\,|\,,$$

where $b = 890$ Å $(=d)$ and $a = \tau b$. A close inspection of this structure reveals that it can be considered to be a one-dimensional *periodic* lattice of the unit *abaab* with randomly placed substitutional defects of the form *aab*. Motivated by this observation we provide in Fig. 5 the Raman scattering spectra of a periodic *abaab* superlattice with Γ equal to (a) 0.02; (b) 0.2; and (c) 0.7 meV. We also give the corresponding plasmon dispersion relation as an inset in Fig. 5(c). Values of all the parameters are the same as those used in Figs. 1–4. A comparison of Figs. 2 and 5 clearly shows that the short-range correlation effects arising from *abaab* units dominate the quasiperiodic spectra, since Figs. 2(a), 2(b), and 2(c) look very similar (even quantitatively) to Figs. 5(a), 5(b), and 5(c), respectively. This is particularly true for higher values Γ (≥ 0.2 meV). Thus it is suggestive to think of the Fibonacci superlattice as a partially ordered layer lattice with no long-range positional order, but with strong short-range order which dominates the plasmon structure.

In conclusion, it is thus clear that one wants to make structures with very small Γ to be able to see distinct effects on plasmons due to aperiodic structures. There are a number of factors contributing to Γ—impurity scattering, variation in the electron density, fluctuation in the layer size, and photon-decay effects being the important ones at low temperatures. Even if one can completely eliminate charged-impurity scattering effects,[17] variations in the electron density and well size from layer to layer make Γ greater than 0.05 meV. If, in addition, one takes into account photon-extinction effects[1] it is unlikely to have a situation with Γ less than 0.1 meV ($\Gamma \cong 0.2$–0.7 meV is more realistic). Given these resolution restrictions, we propose experiments with high N_0 and d (to minimize density and size-fluctuation effects) at high values of q to observe aperiodic effects in superlattice plasmons. We emphasize that a Raman scattering experiment will show differences between the plasmon spectra in periodic and aperiodic structures (as is clear from Figs. 1–5), but the interpretation of such spectra in terms of the physics of quasiperiodic systems will be difficult in view of broadening and finite-size effects. One can, of course, directly compare our theoretical spectra with the experimental ones, and obtain an empirical value for Γ. (This is particularly true for aperiodic systems where the mode structure is very sensitive to small changes in Γ provided it is not too large.) Although it is possible to see some aspects of the mode structure from frequency-scan experiments (Figs. 1–5), a much better experiment is to obtain the Raman spectra in $\mathbf{k}$ space (for fixed ω) where incommensuration effects show up more prominently as has been emphasized in our earlier paper.[5] That, however, is more difficult from an experimental viewpoint. We hope that our work will motivate experimentalists to look for the specific effects predicted in this paper.

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[14]As we show in Ref. 5, the choice of z_j periodic and n_j random or aperiodic allows one to map the plasmon problem directly onto a known Anderson or Aubry model. Whereas the current choice (i.e., z_j random or aperiodic and $n_j = N_0$, a constant) introduces randomness or quasiperiodicity through the exponential function, $\exp(-q\,|z_j - z_{j'}|)$ in Eq. (1): there is no direct known analog in the literature on random or aperiodic systems in this case.

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