Evolution and splitting of plasmon bands in metallic superlattices

S. H. Mahmood, A. Malkawi, and I. Abu-Aljarayesh
Department of Physics, Yarmouk University, Irbid, Jordan
(Received 28 December 1988)

We have derived the dispersion relation for the collective excitations of an infinite superlattice consisting of alternating layers of four different materials, using a simple local theory and neglecting the effects of retardation. The plasmon bands were calculated for several special cases. The number of bands was found to be equal to the number of different materials that make up the superlattice. The band structure and the band gaps were found to be sensitive to the relative thickness of insulating layers that separate two metallic layers in the superlattice.

I. INTRODUCTION

Metallic superlattices have recently generated considerable interest[1] because of the possibility of producing artificial periodic structures with novel magnetic[2–4] and superconductivity[5] properties, and of the feasibility of applying them technologically.[6–8]

Recently, the collective spin-wave[9,10] and plasmon excitations[11,12] of perfect superlattices consisting of alternating layers of two different materials (one or both of which are metals) have been studied theoretically. It was illustrated that the elementary excitations at the interfaces between adjacent layers couple through a macroscopic field to form bands of collective excitations of the whole structure.

In this paper we will use a simple local theory, neglecting the effect of retardation, to investigate the plasmon bands of superlattices, consisting of alternating layers of four different materials. This study will shed light on the effects of oxide layers that may have developed between the metal layers during the preparation process. It will also give information about the effects of introducing periodic semiconducting and/or insulating layers on the band structure of plasmons, and hence give the possibility of fine tuning the band structure and the band gaps of such systems.

The organization of this paper will be as follows. In Sec. II, we derive the dispersion relation for a general superlattice consisting of alternating layers of four different materials. In Sec. III, we discuss the problem of plasmon bands in three different special cases: (a) the plasmon bands for a two-material superlattice, (b) for a three-material superlattice, and (c) for a four-material superlattice.

II. DISPERSION RELATION OF PLASMON BANDS IN A SUPERLATTICE CONSISTING OF ALTERNATING LAYERS OF FOUR DIFFERENT MATERIALS

Figure 1 shows a superlattice constructed of four different materials A, B, C, and D, characterized by the thicknesses and the dielectric constants d_i and $\epsilon_i(\omega)$, where $i=1,2,3,4$. The period of the superlattice $L=\sum_i d_i$.

A given isolated film is capable of supporting bulk as well as surface modes.[13,14] As the different layers are brought together to form the superlattice structure shown in Fig. 1, the interface modes between the various layers couple to form bands of plasmons characteristic of the whole superlattice. At these plasmon frequencies, none of the constituents has a vanishing dielectric constant, i.e., $\epsilon_i(\omega)\neq 0$. Thus, the electrostatic potential $\phi(\mathbf{r},t)$ satisfies Laplace's equation everywhere:

$$\nabla^2\phi(\mathbf{r},t)=0 , \tag{1}$$

and must satisfy the appropriate boundary conditions. The in-plane (x-y plane) isotropy suggests that the potential can be written as[11]

$$\phi(\mathbf{r},t)=\phi(z)e^{i(kx-\omega t)} , \tag{2}$$

with $\mathbf{k}$ taken parallel to the x axis for convenience.

Thus Eq. (1) reduces to the one-dimensional problem

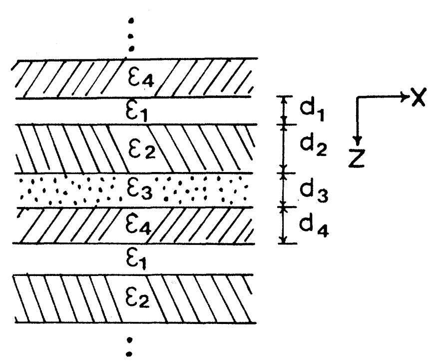

FIG. 1. A schematic diagram of an infinite superlattice consisting of alternating layers of four different materials A, B, C, and D.

$$\left[\frac{d^2}{dz^2}-k^2\right]\phi(z)=0 . \tag{3}$$

The periodicity along the z axis requires that the solution of Eq. (3) should form Bloch waves with respect to translations perpendicular to the interfaces. Thus, we can write

$$\phi(z)=U_q(z)e^{iqz} , \tag{4}$$

where $U_q(z)$ has the periodicity of the superlattices such that

$$U(z)=U(z+nL) , \tag{5}$$

n being an integer. Thus, the general solution of $U_q(z)$ in the various layers of the nth period is

$$U_q(z)=e^{-iq(z-nL)}\times\begin{cases} A_+e^{k(z-nL)}+A_-e^{-k(z-nL)}, & nL+d_1\geq z\geq nL \\ B_+e^{k(z-nL-d_1)}+B_-e^{-k(z-nL-d_1)}, & nL+d_1+d_2\geq z\geq nL+d_1 \\ C_+e^{k(z-nL-d_1-d_2)}+C_-e^{-k(z-nL-d_1-d_2)}, & (n+1)L-d_4\geq z\geq nL+d_1+d_2 \\ D_+e^{k[z-(n+1)L+d_4]}+D_-e^{-k[z-(n+1)L+d_4]}, & (n+1)L\geq z\geq (n+1)L-d_4 . \end{cases} \tag{6}$$

From the continuity of $\phi(z)$ across the boundaries between the different regions, we obtain the following equations.

At $z=nL$,

$$A_++A_--D_+e^{kd_4}e^{-iqL}-D_-e^{-kd_4}e^{-iqL}=0 ; \tag{7}$$

at $z=nL+d_1$,

$$A_+e^{kd_1}+A_-e^{-kd_1}-B_+-B_-=0 ; \tag{8}$$

at $z=nL+d_1+d_2$,

$$B_+e^{kd_2}+B_-e^{-kd_2}-C_+-C_-=0 ; \tag{9}$$

and at $z=nL+d_1+d_2+d_3$,

$$C_+e^{kd_3}+C_-e^{-kd_3}-D_+-D_-=0 , \tag{10}$$

where A_+, A_-, B_+, B_-, C_+, C_-, D_+, and D_- are complex numbers. The continuity of the normal component of the displacement vector $\mathbf{D}$ across the boundaries leads to the following four equations:

$$A_+\epsilon_1-A_-\epsilon_1-D_+\epsilon_4e^{-iqL}e^{kd_4}+D_-\epsilon_4e^{-iqL}e^{-kd_4}=0 , \tag{11}$$

$$A_+\epsilon_1e^{kd_1}-A_-\epsilon_1e^{-kd_1}-B_+\epsilon_2+B_-\epsilon_2=0 , \tag{12}$$

$$B_+\epsilon_2e^{kd_2}-B_-\epsilon_2e^{-kd_2}-C_+\epsilon_3+C_-\epsilon_3=0 , \tag{13}$$

and

$$C_+\epsilon_3e^{kd_3}-C_-\epsilon_3e^{-kd_3}-D_+\epsilon_4+D_-\epsilon_4=0 . \tag{14}$$

By setting the appropriate 8×8 determinant [formed from Eqs. (7)–(14) to zero], the following implicit dispersion relation for the bulk modes of the superlattice was obtained:

$$(\epsilon_1^2\epsilon_3^2+\epsilon_2^2\epsilon_4^2)\prod_{i=1}^{4}\sinh(kd_i)+2\left[\prod_{i=1}^{4}[\epsilon_i\cosh(kd_i)]-\cos(qL)\prod_{i=1}^{4}\epsilon_i\right]+\tfrac{1}{4}\sum_{i\neq j\neq l\neq m}^{4}J_{ijlm}=0 , \tag{15}$$

where

$$J_{ijlm}=\epsilon_i\epsilon_j(\epsilon_m^2+\epsilon_l^2)\cosh(kd_i)\times\cosh(kd_j)\sinh(kd_m)\sinh(kd_l) . \tag{16}$$

In what follows, we consider some numerical examples and special cases of this dispersion relation. The dielectric functions of the metal constituents will be considered to follow the free-electron-gas model, namely

$$\epsilon(\omega)=1-\omega_p^2/\omega^2 , \tag{17}$$

where ω_p is the plasmon energy of the metal under consideration.

III. DISCUSSION AND CONCLUSIONS

In this section we discuss some examples and special cases in which the Al and Mg will be taken as the metallic layers and Al_2O_3 ($\epsilon=3$ as given in Ref. 11) and vacuum ($\epsilon=1$) will be taken as the insulating layers.

A. Metallic superlattices of two different materials

The dispersion relation for this superlattice can be obtained from Eq. (15) by setting $d_2=d_4=0$, and is found to be

$$\left[1+\left[\frac{\epsilon_1(\omega)}{\epsilon_3(\omega)}\right]^2\right]\sinh(kd_1)\sinh(kd_3)$$
$$+2\frac{\epsilon_1(\omega)}{\epsilon_3(\omega)}[\cosh(kd_1)\cosh(kd_3)-\cos(qL)]=0 \ . \qquad (18)$$

This result is consistent with previously published results.[11,12] Figure 2 shows the plasmon bands of Al-Mg superlattices with equal layer thicknesses of Al and Mg, and with plasmon energies for the two constituents of $\omega_p(\mathrm{Al})=15$ eV and $\omega_p(\mathrm{Mg})=10$ eV. This figure shows that the upper branch of the band (with $qL=\pi$) starts at $\omega_p(\mathrm{Al})$ and decreases asymptotically towards the value of the interface plasmon between the Al and Mg layers which has the value of $\{[\omega_p^2(\mathrm{Al})+\omega_p^2(\mathrm{Mg})]/2\}^{1/2}$. The lower branch (also with $qL=\pi$) starts at $\omega_p(\mathrm{Mg})$ and increases smoothly towards the asymptotic value of the upper branch. This apparently single band is actually two bands joining at the branch with $qL=0$. If the two layers become unequal, splitting of the two bands is then observed as in Ref. 11.

B. Metallic superlattices of three different materials

The dispersion relation for this superlattice was initially solved explicitly by setting up the appropriate 6×6 determinant and the solution was found to be

$$2\left[\prod_{i=1}^{3}\epsilon_i\cosh(kd_i)-\cos(qL)\prod_{i=1}^{3}\epsilon_i\right]+\frac{1}{2}\sum_{i\neq j\neq l}J_{ijl}=0 \ , \qquad (19)$$

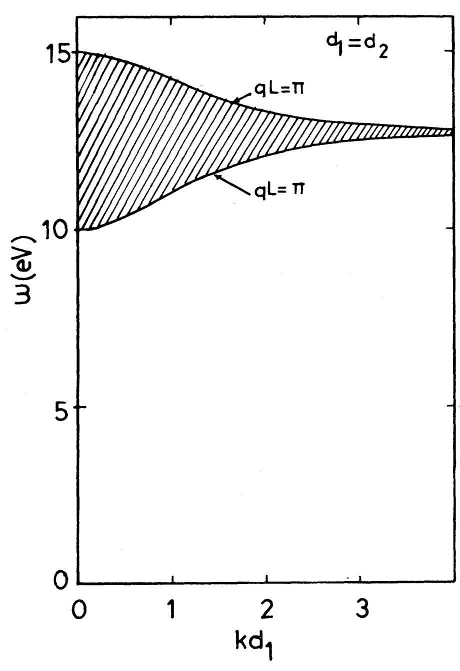

FIG. 2. Dispersion relation for the Al/Mg superlattice with equal layer thicknesses and with $\omega_p(\mathrm{Mg})=10$ eV and $\omega_p(\mathrm{Al})=15$ eV.

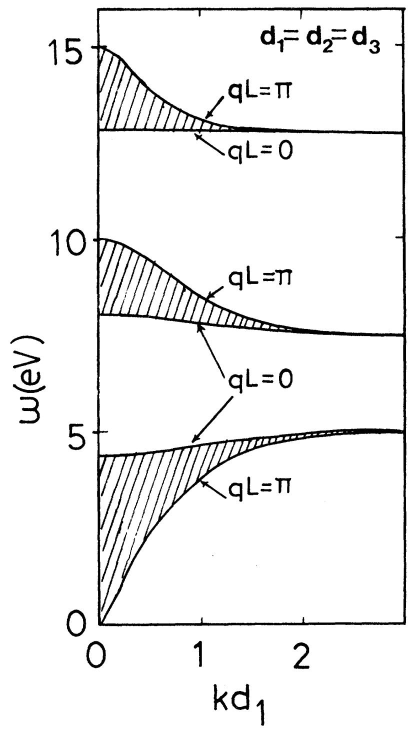

FIG. 3. Dispersion relation for the Al/Mg/Al_2O_3 superlattice with equal layer thicknesses and with $\epsilon(Al_2O_3)=3$.

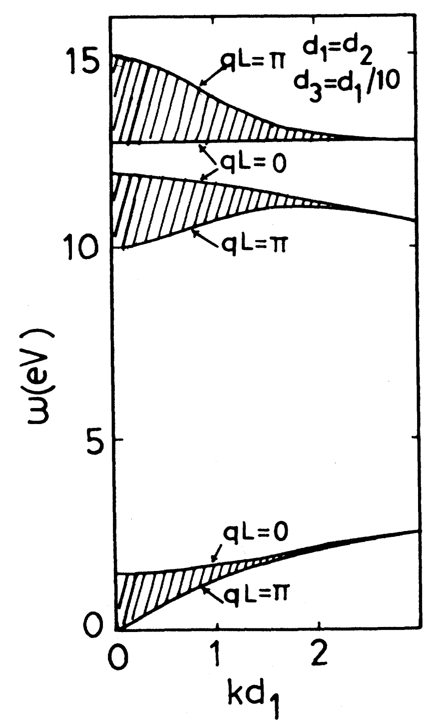

FIG. 4. Dispersion relation for the superlattice of Fig. 3 with $d_1=d_2$ and $d_3=d_1/10$.

where

$$J_{ijl}=\epsilon_i(\epsilon_j^2+\epsilon_l^2)\cosh(kd_i)\sinh(kd_j)\sinh(kd_l) \ . \quad (20)$$

This dispersion relation was also obtained from Eq. (15) by setting any of the four layer thicknesses equal to zero. Figure 3 shows that there are three well-separated bands for the superlattice Al/Mg/Al_2O_3 with equal layer thicknesses. The branch with $qL=\pi$ for the uppermost band starts at ω_p(Al) and decreases asymptotically towards the value of the interface plasmon between Al and Mg. The asymptotic value of this band is associated with the interface plasmons between Al and Mg. The middle band starts with $qL=\pi$ at ω_p(Mg) and decreases asymptotically towards the value of the interface plasmon between Al and Al_2O_3. Finally, the lower band reaches an asymptotic value which is equal to the value of the interface plasmon between Mg and Al_2O_3.

As the layer thickness of the Al_2O_3 is decreased, the band gap between the upper and the middle bands decreases and the lower band is pushed downward in energy. This behavior is consistent with the limiting process as the thickness of Al_2O_3 approaches zero. Figure 4 shows the band structure of the same superlattice corresponding to Fig. 3 but with the thickness of the oxide layer reduced to 10% of its previous value.

C. Metallic superlattices of four different materials

Here we discuss numerically the plasmon bands of the superlattice Mg/vacuum/Al/Al_2O_3. Without any loss of generality, the vacuum layer was chosen arbitrarily as one of the four constituents of the superlattice.

Figure 5 shows the plasmon bands for the superlattice with equal layer thicknesses of the four constituents. The upper band starts at ω_p(Al) and the fairly narrow lower band starts at $\omega=0$. Each of these two bands is surrounded by two modes with $qL=0$ and $qL=\pi$. A relatively wide middle band is surrounded by the two modes each with $qL=\pi$. This band actually consists of two bands merging together (the modes with $qL=0$ in these two bands coincide).

In this superlattice we have four different interface plasmons: the Mg/vacuum interface plasmon with energy $\omega_p(\mathrm{Mg})/\sqrt{2}=7.1$ eV, the Al/vacuum plasmon with energy $\omega_p(\mathrm{Al})/\sqrt{2}=10.6$ ev, the Al/Al_2O_3 plasmon with energy $\omega_p(\mathrm{Al})/\sqrt{4}=7.5$ eV, and finally the Mg/Al_2O_3 plasmon with energy $\omega_p(\mathrm{Mg})/\sqrt{4}=5$ eV. In Fig. 5 we notice that the bands approach the above four energies

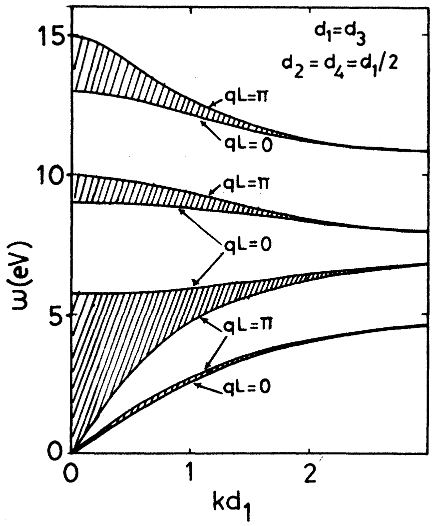

FIG. 6. Dispersion relation for the superlattice of Fig. 5 with $d_1=d_3$ and $d_2=d_4=d_1/2$.

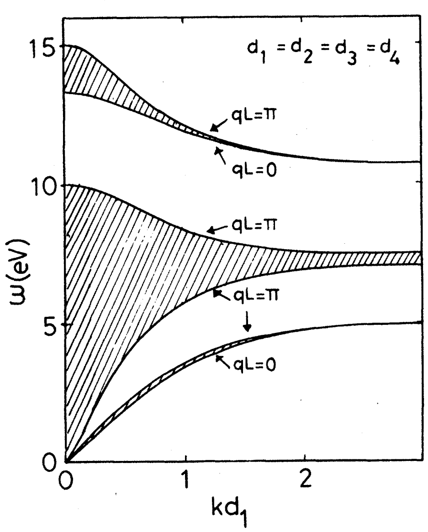

FIG. 5. Dispersion relation for the Mg/vacuum/Al/Al_2O_3 superlattice with equal layer thicknesses.

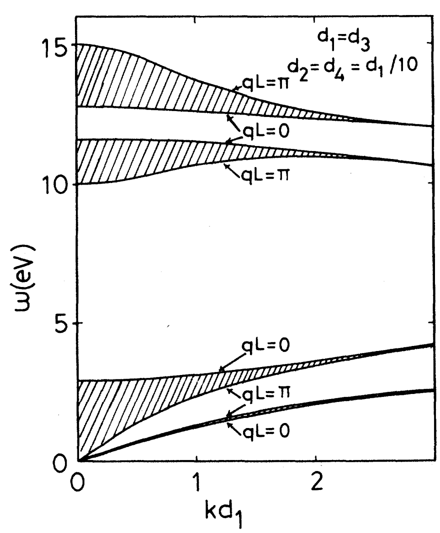

FIG. 7. Dispersion relation for the superlattice of Fig. 6 with $d_2=d_4=d_1/10$.

asymptotically, indicating that these bands correspond to the coupling between the interface plasmons at the adjacent boundaries.

As the layer thicknesses of the two insulators are decreased, the middle wide band splits into two bands and the $qL=0$ branches appear as shown in Fig. 6. These two bands become further apart and the lower bands decrease in energy as we further decrease the layer thicknesses of the insulators as shown in Fig. 7. We also notice from this figure that as we decrease the layer thicknesses of the insulators the band gap between the two upper bands decreases and the band structure resembles that of the superlattices with two different materials, indicating that there might be a coupling between the plasmons in the Al and Mg layers across the vacuum layer, a phenomenon reminiscent of the tunneling effect.

[1]I. Schuller and C. Falco, in *VLSI Electronics*, edited by N. G. Einspruch (Academic, New York, 1982), Vol. 4.
[2]J. Kwo, M. Gyorgy, D. McWhan, M. Hong, F. D. Salvo, C. Vellier, and J. Bower, Phys. Rev. Lett. **55**, 1402 (1985).
[3]I. Schuller and M. Grimsditch, J. Appl. Phys. **55**, 2491 (1984).
[4]E. Gyorgy, J. Dillon, D. McWhan, L. Rupp, Jr., L. Testardi, and P. Flanders, Phys. Rev. Lett. **45**, 57 (1980).
[5]C. Chun, G. Zheng, J. Vincent, and I. Schuller, Phys. Rev. B **29**, 4915 (1984).
[6]E. Spiller, Appl. Opt. **15**, 2333 (1976).
[7]T. Barbee, in *Low Energy X-ray Diagnosis*, edited by D. Attwood and B. Hencker (AIP, New York, 1981), pp. 57 and 131.
[8]A. Sakena and B. Schoenborn, Acta Crystallogr. Sect. A **33**, 805 (1977).
[9]P. Grunberg and K. Mika, Phys. Rev. B **27**, 2955 (1983).
[10]R. E. Camely, T. S. Rahman, and D. L. Mills, Phys. Rev. B **27**, 261 (1983).
[11]R. E. Camely and D. L. Mills, Phys. Rev. B **29**, 1695 (1983).
[12]G. Giuliani, J. Quinn, and R. Wallis, J. Phys. (Paris) Colloq. **45**, C5-285 (1984).
[13]E. Stern and R. Ferrell, Phys. Rev. **120**, 130 (1960).
[14]R. Ritchie, Phys. Rev. **106**, 874 (1957).
[15]H. Raether, *Excitation of Plasmons and Interband Transitions by Electrons* (Springer-Verlag, Berlin, 1980).