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Plasmons in amorphous multilayer films

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We show, with use of a field-emission scanning transmission electron microscope, changes in the plasmon region from thin films of silicon in a cobalt matrix. These effects are consistent with the dielectric-response theory developed by Howie.

It is now possible, using the field-emission scanning transmission electron (STEM), to record electron-energy-loss spectra from nanometer-sized regions. Apart from high spatial resolution microanalysis using characteristic inner-shell signals, there is some interest in detecting changes in the low-loss region that might be due to quantum size effects. In particular Batson[1] showed peaks corresponding to the allowed modes of a free-electron gas in small aluminum spheres and also showed the interface plasmons between the dielectric oxide surface layer and the metal sphere. A more detailed analysis from spheres on various substrates has been given by Wang and Cowley.[2-4]

In this paper we report the observation of low-loss spectra (up to 30 eV loss) as a probe is moved across a 10-Å silicon layer in a cobalt-silicon multilayer film. We show that the changes in the spectrum can be modeled by the dielectric-response-function theory of Howie[5] and Garcia-Molina *et al.*[6]

Multilayers of cobalt and silicon were prepared by successive UHV evaporations at a rate of 0.5 Å/sec with initial deposition onto the [100] surface of a silicon substrate. Cross-sectional specimens were prepared using the technique of Bravman and Sinclair[7] for examination by scanning transmission electron microscopy. A standard dark-field image taken using a probe of diameter 10 Å, a divergence semiangle of 4 mrad, and a collection semiangle of 6 mrad is shown in Fig. 1. The thin silicon layers appear as dark areas as they scatter less than the cobalt matrix. The specimen was estimated to have a thickness of 350±50 Å by taking a ratio of the total inelastic scattering to the zero-loss peak.[8] Figure 2 shows the geometry of the speciment with respect to the electron beam.

An energy-loss spectrum from the silicon layer 10 Å wide is shown in Fig. 3(a) (solid line) and compared with a spectrum taken from a similar thickness of amorphous silicon (dashed line). Note the broadening of the plasmon peak to about 8.5 eV from 4.0 eV and the shift in the position of the maximum by about 2.0 eV. At first it was thought that this might be a manifestation of quantum size effects on the plasmon modes in a thin layer. This should result in quantization of the component of momentum normal to the layer and the plasmon dispersion should be that appropriate for a two-dimensional electron gas. Preliminary calculations showed that if the layer thickness was 10–20 Å, the component of momentum perpendicular to the interface would be 0.3–0.6 Å^{-1} and these effects would not be observable if the width of the plasmon (due to finite lifetime) is of order 1.5 eV.

For comparison, Fig. 3(b) shows an energy-loss spectrum from pure cobalt (solid line) and the spectrum from the thin silicon layer (dashed line). The cobalt peak is much broader, half-width 15 eV, and the maximum is at an energy of 20.5 eV, which is 1.8 eV higher than the energy of the maximum of the plasmon from the thin silicon layer. At first sight the features in the spectrum from the thin layer are closer to those of pure cobalt; although it might be thought that this is a consequence of beam spreading, calculations show that beam broadening is negligible for this thickness of silicon.[9]

Recently Howie[5] and Garcia-Molina *et al.*[6] have published expressions for the energy-loss spectra from a system of three layers in terms of the frequency-dependent dielectric constant of each medium. They derive their ex-

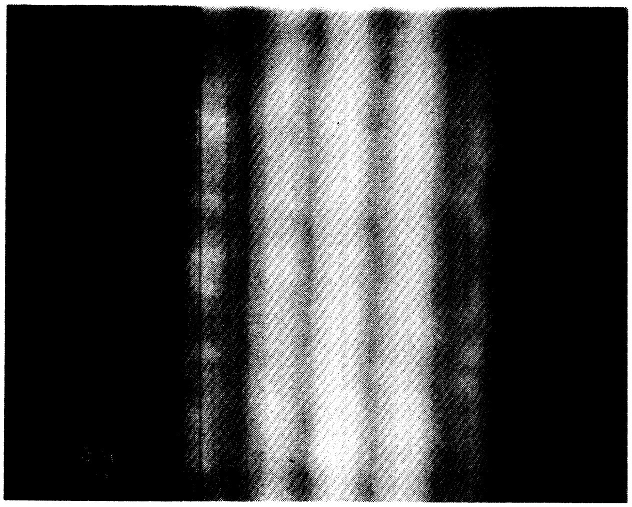

FIG. 1. Dark-field image of cobalt-silicon multilayers.

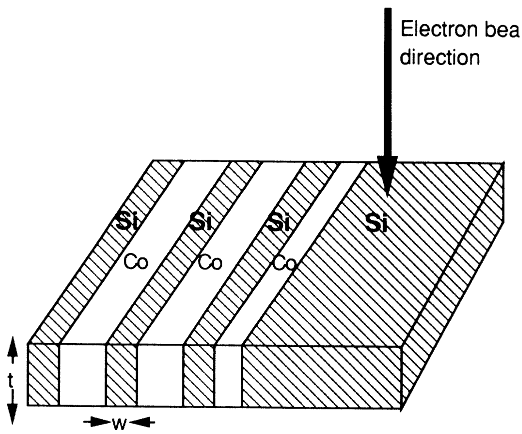

FIG. 2. Schematic illustrating the geometry of the specimen with respect to the electron beam.

pression from the classical electrodynamics of a moving point charge. Programs have been written for calculating observed energy-loss spectra as a probe in a STEM is moved across a surface or interface. Batson[10] has applied the theory to explaining the spectrum from a beam at grazing incidence to a film of aluminum oxide on aluminum. Results of the application of the theory to energy-loss spectra from a Si/SiO$_2$ interface, the (110) surface of GaAs, and the (100) surface of a MgO cube have been reported by Howie and Milne.[11] They managed to reproduce all the features of the low-loss spectra, which in some cases might have been confused with surface or interface states. The theory has been extended to cylindrical interfaces by Walsh,[12] who showed that the calculations reproduced the shape of a spectrum from a beam passing through a hole drilled in AlF$_3$.

The differential cross section for a beam incident at the center of a layer, dielectric constant ϵ', surrounded by material of dielectric constant ϵ is

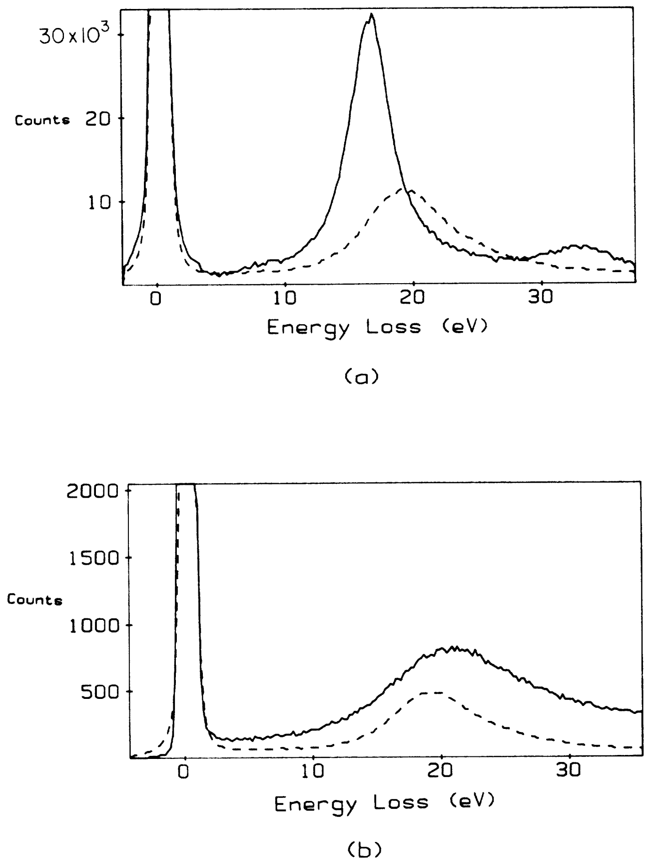

FIG. 3. (a) Electron-energy-loss spectrum taken from Si layer 10 Å wide (dashed line) compared with an energy-loss spectrum taken from bulk silicon of the same thickness (solid line). The spectra have been scaled so that the zero-loss intensities are identical. (b) Comparison of the energy-loss spectrum from 10-Å-wide silicon layer (dashed line) and the spectrum from cobalt (solid line).

$$\frac{d^2I}{dq\,dE}=\frac{e^2}{2\pi^2\epsilon_0\hbar^2v^2k}\left\{-\frac{[(\epsilon^2+\epsilon'^2)\cosh(Kw)+2\epsilon\epsilon'\sinh(Kw)+(\epsilon'^2-\epsilon^2)]}{\epsilon'[(\epsilon^2+\epsilon'^2)\sinh(Kw)+2\epsilon\epsilon'\cosh(Kw)]}\right\}, \tag{1}$$

where

$$K^2=q^2+E^2/(\hbar v)^2 .$$

q is the momentum transfer parallel to the specimen surface, v is the fast electron velocity, E the energy loss, and w the width of the layer. Both the real and imaginary parts of the dielectric constant are required in the theory. These were obtained by a Kramers-Kronig analysis of spectra from pure silicon and pure cobalt using the Fourier-transform method of Johnson.[13] The results from the theory are compared with the experiment in Fig. 4. Both the shift and broadening of the plasmon peak are faithfully reproduced. This illustrates that the

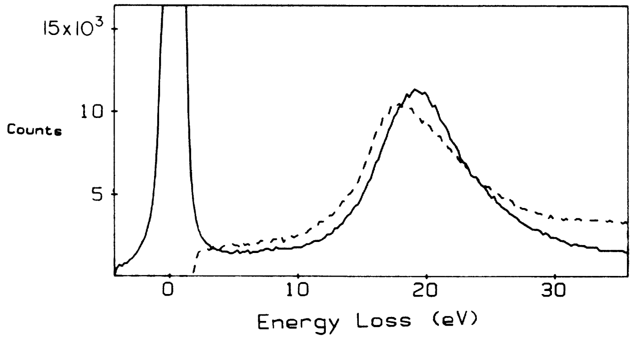

FIG. 4. Electron-energy-loss spectrum from silicon layer 10 Å wide (solid line) compared with calculated spectrum (dashed line).

change in shape of the plasmon peak for a thin silicon layer can be explained by the dielectric response theory.

In conclusion, we have shown that the energy-loss spectrum from a thin layer of silicon in a cobalt-silicon multilayer structure can be explained using the classical dielectric theory of Howie[5] and Garcia-Molina *et al.*[6]

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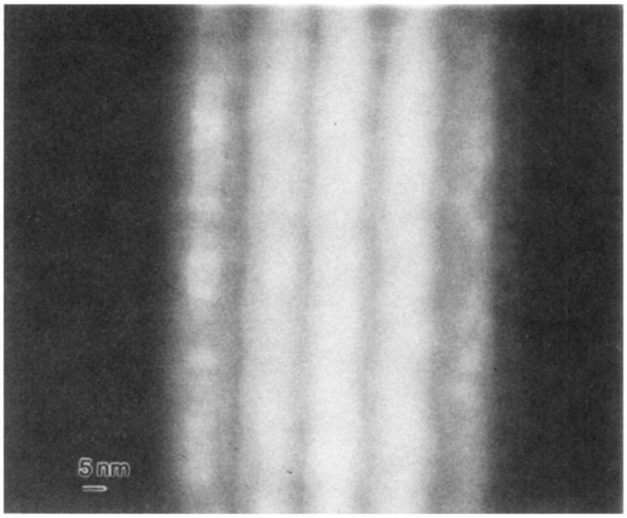

FIG. 1. Dark-field image of cobalt-silicon multilayers.