Observation of Double-Plasmon Excitation in Aluminum

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We present experimental evidence confirming the existence of the theoretically proposed double-plasmon—excitation process in solid-state plasmas. Preliminary measurements of the relative probability of single- and double-plasmon excitation in aluminum show good agreement with theory.

The mean free path for the second-order process of double-plasmon excitation in a free-electron gas has recently been calculated by Ashley and Ritchie.[1] Previous treatments of the interaction of fast electrons with a free-electron plasma ignored the possibility of spontaneous multiple-plasmon excitation and considered that only a single plasmon of energy $\hbar\omega_p$ (ω_p is the plasma frequency) is excited. Multiple-plasmon peaks have been observed in electron—energy-loss spectra from a wide range of materials, but these have always been entirely attributed to plural-scattering single-plasmon processes. These mask the multiple-plasmon single-scattering contributions which would occur at the same energy.

As shown by Ashley and Ritchie, the mean free paths for the single- and double-plasmon processes are related by

$$\lambda_2^{-1} = (9k_c^5\hbar/2^7\pi^2 nm\omega_p)\lambda_1^{-1},$$

where k_c is the cutoff wave number. It readily follows that a two-parameter Poisson distribution is now required to describe the zeroth, first, second, etc. plasmon-loss probabilities in the energy-loss spectrum. As a function of plasma thickness t these are given by

$$P_0(t) = \exp(-t/\lambda),$$

$$P_1(t) = (t/\lambda_1)\exp(-t/\lambda),$$

$$P_2(t) = \tfrac{1}{2}(t/\lambda_1)^2\exp(-t/\lambda) + (t/\lambda_2)\exp(-t/\lambda),$$

where

$$\lambda^{-1} = \lambda_1^{-1} + \lambda_2^{-1}.$$

In the expression for $P_2(t)$ the first term is due to single-plasmon double-scattering and the second due to double-plasmon single-scattering processes.

Representing the free electrons in a thin crystal as a typical solid-state plasma and taking the value $k_c \approx \omega_p/v_{\mathrm{F}}$ (v_{F} being the Fermi velocity), Ashley and Ritchie find that, at a thickness equal to λ_1, approximately 8% of the second plasmon-loss intensity arises from the double-plasmon contribution. Thus at small thicknesses the influence of the double-plasmon term should be detectable experimentally.

The present experiments involve the careful measurement of the zeroth, first, and second plasmon-peak intensities of the fast-electron energy-loss spectrum as a function of plasma thickness in the hope of identifying this double-plasmon contribution to $P_2(t)$. The technique is an extension of that previously described[2] for the measurement of λ_1.

An electron-microscope system with electronic recording and energy-analysis facilities[3] is used to examine wedge-shaped crystals of aluminum. The crystal is first oriented by means of a goniometer stage to an approximate "one-beam condition" in which no Bragg-diffracted beam is strongly excited. The resulting image of the crystal, with no objective aperture in place, is then scanned perpendicular to the edge of the wedge so that the number of transmitted electrons which have lost energy $n\hbar\omega_p$ ($n = 0, 1, 2$) is recorded as a function of crystal thickness. The energy window was chosen so as to include the full width of the plasmon-loss peaks as determined from an experimental differential energy spectrum from the same crystal.

Figure 1 shows typical results for 67-keV incident electrons. The ordinate (intensity) is in arbitrary units. The thickness-calibration scale for the abscissa was obtained by scanning a set of Pendellösung fringes in a dark-field image formed with the (200) Bragg reflection satisfied. By comparing the elastically transmitted electron contribution to the Pendellösung-image profile with a fifteen-beam dynamical calculation (including absorption), the thickness of the crystal at each point of the scan could be determined. Thus the experiment consists of four successive scans across the same line in the image: the first on a (200) dark-field image with the crystal oriented to the exact Bragg position and in which only the elastically scattered electrons are

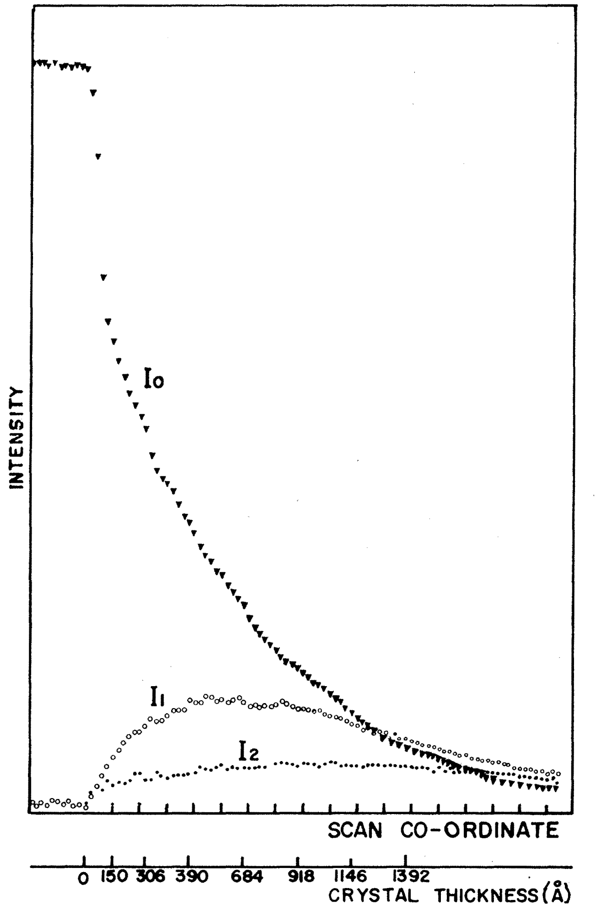

FIG. 1. Elastic (I_0), first plasmon-loss (I_1), and second plasmon-loss (I_2) transmitted-electron intensity as a function of crystal thickness.

counted, and the other three with the crystal tilted to an approximate one-beam condition and the energy analyzer set to record successively the zeroth, first, and second plasmon-loss electrons.

In analyzing these experimental results it is necessary to include a contribution to the overall mean free path due to scattering outside the experimentally defined "windows." This could arise from other energy-loss processes or large-angle elastic scattering outside the microscope-objective pole piece. Thus the one-plasmon-loss intensity should be given by $I_1(t)=(t/\lambda_1)\times\exp(-t/\lambda_e)$, where $\lambda_e^{-1}=\lambda_1^{-1}+\lambda_2^{-1}+\lambda_0^{-1}$ with λ_0 the additional contribution for scattering outside the plasmon windows. Then at $t=\lambda_e$, $I_1(t)$ has a maximum value and the value of λ_e/λ_1 can be obtained from the relation $\lambda_e/\lambda_1=I_1(\lambda_e)/I_0(\lambda_e)$.

Figure 2 shows the first plasmon-loss plot after the abscissa has been rescaled so that it appears as it would for a crystal of uniformly in-

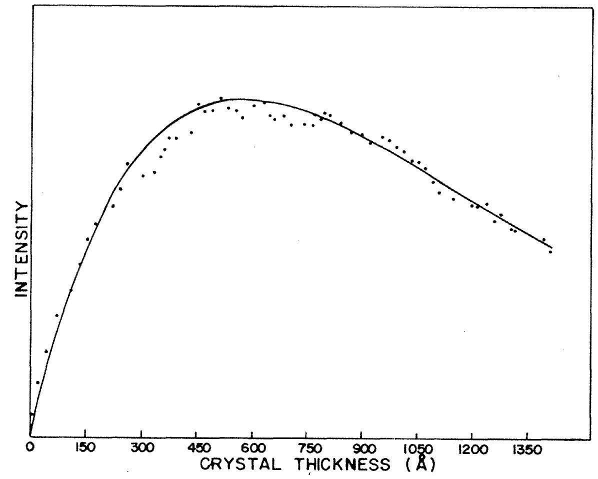

FIG. 2. Thickness-rescaled first plasmon-loss intensity as a function of crystal thickness. The continuous line shows the curve of best fit of the function $I_1(t)$.

creasing thickness. A least-squares fit by the function $I_1(t)$ is also indicated using the value $\lambda_e/\lambda_1=0.39$ as obtained from the maximum of the experimental plot. The curve of best fit yields the values $\lambda_e=575$ Å and $\lambda_1=1490$ Å.

In Fig. 3 the thickness-rescaled, second-plasmon-loss experimental data are plotted and compared with the function (dashed curve),

$$I_2'(t)=\tfrac{1}{2}(t/\lambda_1)^2\exp(-t/\lambda_e),$$

which would apply if double-scattering single-plasmon excitation is the only contribution to the $2\hbar\omega_p$-loss peak. A much better fit is obtained

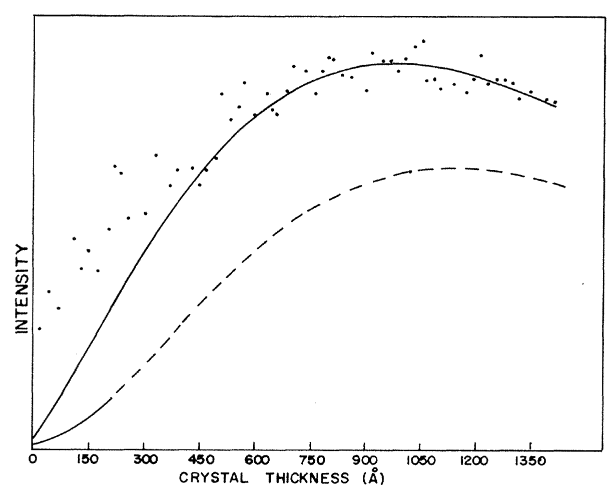

FIG. 3. Thickness-rescaled second plasmon-loss intensity as a function of crystal thickness. The dashed line is the theoretical curve assuming only single-plasmon excitation, while the continuous curve includes the double-plasmon contribution.

(continuous line) using

$$I_2(t)=\tfrac{1}{2}(t/\lambda_1)^2\exp(-t/\lambda_e)+(t/\lambda_2)\exp(-t/\lambda_e),$$

with $\lambda_2=7.41\lambda_1$, the value from the Ashley and Ritchie result using $k_c=1.5$ Å^{-1}. This k_c value is approximately the mean of several experimental results.[4] The value for λ_0 is then 1020 Å.

The fit between theory and the experimental data is not so good at low thicknesses. This is probably because of the presence of oxide and contamination layers, which are more important in the low-thickness region. An analysis of this problem and also details of the errors inherent in the thickness rescaling, the one-beam approximation, and the large-angle scattering will be reported in a later paper. While these errors affect slightly the numerical values for the individual parameters, the present results apparently confirm the existence of the double plasmon and show good agreement with the theoretical value of the ratio λ_2/λ_1. It should be noted, however, that the theoretical λ_2/λ_1 is extremely sensitive to the value assumed for k_c (being proportional to k_c^5) and a sufficiently accurate value of this parameter is not available. The value of λ_1 obtained here is much larger than previously reported by several authors. This is because of the previous use of a single-parameter Poisson distribution which neglects the double plasmon and other processes.

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New Mechanism for Internal Friction*

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Simultaneous measurements of damping and elastic modulus of copper as affected by electron irradiation have been made. These data do not follow the standard analysis, using the Granato-Lücke theory for damping, in which point defects, created by irradiation, are presumed to act as firm pinning points on dislocation lines. It is proposed instead that these defects are *dragged* along by the dislocation line moving under oscillating stress. This dragging can lead to an observed "anomalous" peak (initial increase and subsequent decrease) in the decrement as a function of electron dose.

The analyses of a large number of experiments on internal friction in metals have been based, with remarkable success, on a formulation of Koehler,[1] amplified in detail by Granato and Lücke,[2] the G-L theory. This theory, a string model for dislocation bowing under applied stress, is best known for its predictions, at reasonably low frequencies, for the strain amplitude-independent decrement δ and modulus change $\Delta E/E$:

$$\delta=aB\omega\Lambda L^4, \tag{1}$$

$$\Delta E/E=b\Lambda L^2, \tag{2}$$

B is a viscous damping constant, ω is the angular drive frequency, Λ is the total length of dislocation line per unit volume, L is the average length of dislocation between pinning points, and a and b are constants. The L^4 and L^2 dependences in Eqs. (1) and (2) are watermarks of the G-L theory. Irradiation experiments are particularly well suited for testing this L^4-L^2 prediction since the accretion of defects on dislocation lines during bombardment or subsequent annealing provides a controlled method for apparently systematically shortening the loop length L while not affecting any other parameters. If we let

$$Y\equiv\frac{\Delta E/E}{(\Delta E/E)_0}\simeq\frac{\Delta E}{\Delta E_0} \tag{3}$$

and

$$Z\equiv\delta/\delta_0, \tag{4}$$