than 0.5%.

impurities were nonmagnetic, then BCS should apply and the specific-heat jump would be proportional to T_c/T_{cp} , as shown by the dashed line. For magnetic impurities, SBW predicts the solid curve, in excellent agreement with the experiments. The open circles are calorimetric determinations of the specific-heat jump take in this laboratory for La-Gd alloys.^{6,26} Both the specific-heat and critical-field results show the effect but the critical-field data illustrate the point to much higher accuracy.

Values of ρ , H_0 , T_c , $\Gamma/\Delta_p(0)$, and $C_s - C_n \mid_{T=T_c}$ are all summarized in Table II. A complete tabulation of all the data is presented elsewhere.¹³

CONCLUSIONS

Pure thorium is a weak-coupling type-I superconductor with critical-field curves which follow the BCS ²⁶ The data for the highest concentration have not yet been published in Ref. 6.

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Radiative Decay of Coulomb-Stimulated Plasmons in Spheres*

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We have studied the excitation of plasmons by energetic electrons, and their subsequent decay, in small spheres. We estimate that plasma decay light from electron bombardment of small, randomly distributed metal spheres suspended in a dielectric medium may be emitted with intensity comparable with that found in experiments on plasma decay light from metallic slabs.

I. INTRODUCTION

THE characteristic absorption of light by small I metal spheres has recently been studied experimentally by Doremus¹ and theoretically by Kawabata and Kubo.² Doremus's data for the wavelength dependence of the absorption coefficient for 100 Å silver and gold spheres show peaks which are explained on the basis of plasma resonance absorption by the conduction electrons. Kawabata and Kubo have used linear response theory to explain the widths of these resonances in terms of the damping of the plasma oscillations by transfer of energy to single-electron modes. They also take account of radiation damping, and their results show good agreement with Doremus's data.

theory to an accuracy of 0.3%. For additions of Gd

impurity in concentrations up to 0.2%, the samples

remain type-I superconductors which exhibit a Meissner

effect in the gapless regime near T_c as well as the regime where there is a well-defined gap at low tem-

peratures. The AG-SBW theory^{1,12} predicts the critical-

field curves for Th-Gd alloys to an accuracy of better

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The successful explanation of light absorption peaks in terms of collective resonances in a spherical electron gas leads one to speculate about the possibility of stimulating similar resonances by means of energetic, nonrelativistic electrons and observing the radiative decay of these collective states. It is well known that such resonances can be produced in metal foils by electron bombardment and that light from the damping of these resonances can be observed.3 Plasmon decay light from foils was first predicted by Ferrell.⁴ Recent characteristic energy-loss experiments by Fujimoto,

^{*} Research sponsored by the U.S. Atomic Energy Commission under contract with Union Carbide Corporation. † Consultant to Oak Ridge National Laboratory

¹ R. H. Doremus, J. Chem. Phys. 40, 2389 (1964); 42, 414 (1965) ² A. Kawabata and R. Kubo, J. Phys. Soc. Japan 21, 1765

^{(1966).}

⁸ A. L. Frank, E. T. Arakawa, and R. D. Birkhoff, Phys. Rev. **126**, 1947 (1962); E. T. Arakawa, N. O. Davis, L. C. Emerson, and R. D. Birkhoff, J. Phys. Radium **25**, 129 (1964). ⁴ R. A. Ferrell, *ibid.* **111**, 1214 (1958); also, R. H. Ritchie and H. B. Eldridge, *ibid.* **126**, 1935 (1962).

Komaki, and Ishida⁵ involving bombardment of small metal spheres by electrons show that the excitation process may occur with measurable probability.

In what follows we consider a system of small metal spheres embedded at random in a thin dielectric slab, and we inquire whether it is feasible to expect to see light from plasmon decay in the spheres. Since the spheres are considered to be distributed randomly in a transparent medium at an average distance large compared with their radii and with the parameter v/ω_r , where **v** is the velocity of the incident electron and ω_r is the resonant frequency of collective oscillations in the spheres, we may expect that plasma oscillations in different spheres can be considered to occur independently and that only incoherent addition of radiation fields will be necessary if there are many spheres present.

In Sec. II we treat the free electrons in a sphere in the hydrodynamical approximation, and we use this model to quantize the surface plasmon field and to obtain the dispersion relation for surface plasma oscillations. In Secs. III and IV we calculate, in the dipole approximation, the cross section for surface plasmon creation by fast electrons and the radiative decay rate for surface plasmons. Finally, in Sec. V, the complete process consisting of photon decay of a surface plasmon following excitation by a fast electron is considered and an expression for the differential cross section for this process is given.

We also use the present model to calculate the cross section for the process in which a photon excites a surface plasmon and compare our results with the classical Mie theory. This is taken up in Sec. VI.

II. HYDRODYNAMICAL MODEL

In order to carry out the calculations we start with the Bloch linearized, hydrodynamical model of an electron gas.^{6,7} The appropriate equations—(1) the force equation, (2) Poisson's equation, and (3) the continuity equation-are

$$\nabla \frac{\partial}{\partial t} \psi(\mathbf{r}, t) = -\frac{e}{m} \nabla \phi(\mathbf{r}, t) + \frac{\beta^2}{n_0} \nabla n(\mathbf{r}, t), \quad (1)$$

$$\nabla^2 \boldsymbol{\phi}(\mathbf{r}, t) = 4\pi e n(\mathbf{r}, t), \qquad (2)$$

$$\nabla^2 \boldsymbol{\psi}(\mathbf{r}, t) = (\partial/\partial t) n(\mathbf{r}, t) / n_0, \qquad (3)$$

where $\phi(\mathbf{r}, t), \psi(\mathbf{r}, t)$, and $n(\mathbf{r}, t)$ are perturbations in the electric potential, velocity potential, and electronic density, respectively, in the electron gas. n_0 is the electronic density in the undisturbed state of the electron gas, and β is the rms propagation speed of the disturbance through the electron gas. The appropriate

boundary conditions are (i) continuity of the electric potential at the surface of the sphere, (ii) continuity of the normal component of the electric displacement at the surface of the sphere, and (iii) the vanishing of the normal component of the disturbance velocity at the surface of the sphere.

Under these conditions one finds the following dispersion relation for surface plasmons:

$$\frac{l}{l+1} \left(1 - \frac{\omega_p^2}{\omega_t^2} \right) j_{l-1} [iR(\omega_p^2 - \omega_l^2)^{1/2} / \beta]$$

= $\left[1 + \frac{\omega_p^2}{\omega_t^2} \frac{l(\epsilon^0 - 1)}{\epsilon^0(l+1) + l} \right] j_{l+1} [iR(\omega_p^2 - \omega_l^2)^{1/2} / \beta], \quad (4)$

where ω_l is the spherical surface plasma frequency corresponding to a collective oscillation in the angular mode corresponding to the *l*th spherical harmonic, $\omega_p = [4\pi n_0 e^2/m]^{1/2}$ is the volume plasma frequency, R is the radius of the sphere, and ϵ^0 is the dielectric constant of the medium in which the sphere is suspended. $j_l(x)$ is the spherical Bessel function of order l.

In this paper we shall consider only the limiting case $\beta^2 \rightarrow 0$, in which case Eq. (4) reduces to $\omega_l^2 / \omega_p^2 =$ $l/\lceil \epsilon^0(l+1)+l \rceil$, as one may show using the asymptotic expansions of the j_l . Further, we ignore the effect of the dielectric, i.e., take $\epsilon^0 = 1$, for simplicity, and limit ourselves to the dipole approximation l=1. The resulting approximation to the dipole surface plasmon frequency is $\omega_1 = \omega_p / \sqrt{3}$ (cf. Ref. 7). We assume that the dielectric permittivity of the slab is essentially constant and not greatly different from unity over the range of frequencies involved in collective motion of electrons in the spheres. Thus we neglect any screening of the field of the incident electron by the dielectric during its passage through the film, as well as any dynamical response by the dielectric in this same frequency range. Although these approximations limit, somewhat, the range of applicability of the present work, the general form of the results given below should be maintained in a treatment of a more realistic situation; e.g., one would like to treat the effect of a thin dielectric slab, in which plasma spheres are suspended, upon the radiation field from plasmon decay.

The system can be quantized using the linearized Hamiltonian

$$H = -\int d\mathbf{r} \left\{ \frac{1}{2}mn_0 \psi \nabla^2 \psi + \frac{1}{2}e\phi n - \frac{m\beta^2}{2n_0} n^2 \right\}.$$
 (5)

Thus, keeping in mind the approximations discussed above, one has for the dipole term of the operator for electric potential inside the sphere

$$\hat{\phi}(\mathbf{r},t) = -\left(\frac{2\pi\hbar\omega_{1}r^{2}}{3R^{3}}\right)^{1/2} \sum_{s=1}^{3} Y_{1s_{s}}(\Omega) \left(b_{s_{s}} + b_{s_{s}}^{\dagger}\right), \quad (6)$$

where $Y_{1x_s}(\Omega)$ is the real, normalized spherical har-

⁵ F. Fujimoto, K. Komaki, and K. Ishida, J. Phys. Soc. Japan 23, 1186 (1967).
⁶ F. Bloch, Z. Physik 81, 363 (1933).
⁷ H. Jensen, Z. Physik 106, 620 (1937).



monic⁸ corresponding to l=1, and b_{x_s} and $b_{x_s}^{\dagger}$ are, respectively, annihilation and creation operators for surface plasmons oscillating in the x_s direction. In the region r > R, the factor r^2 is to be replaced by R^6/r^4 .

III. CROSS SECTION FOR SURFACE PLASMON CREATION BY FAST ELECTRONS

We wish to calculate the cross section for the process shown in Fig. 1, i.e., the excitation of a surface plasmon in the sphere by an incident electron. We express the wave function of the perturbing electron as a planewave expansion and calculate the interaction Hamiltonian V_{ep} between the electron and the plasma from the equation

$$V_{ep} = -e \int d\mathbf{r} \, \Psi^{\dagger} \Psi \hat{\boldsymbol{\phi}}, \qquad (7)$$

where Ψ is the electron field operator and $\hat{\phi}$ is the operator for perturbation in the electric potential given in (6). In the dipole approximation, the matrix element for the excitation of a plasmon oscillating in the x_{\bullet} direction is

$$\langle \mathbf{k}, s \mid V_{ep} \mid \mathbf{k}' \rangle = i (96\pi^3 e^2 \hbar R \omega_1 / L^6)^{1/2} Y_{1x_s}(\Omega_\kappa) j_1(\kappa R) / \kappa^2,$$
(8)

where $\kappa = \mathbf{k}' - \mathbf{k}$, L^3 is the quantization volume for the plane waves, and $\omega_1 = \omega_p / \sqrt{3}$.

The transition rate is obtained from the Golden Rule, i.e.,

$$w_{s} = (2\pi/\hbar^{2}) \mid \langle \mathbf{k}, s \mid V_{ep} \mid \mathbf{k}' \rangle \mid^{2} \delta(\omega_{\mathbf{k}} + \omega_{1} - \omega_{\mathbf{k}'}), \quad (9)$$

and the total cross section for excitation of a surface plasmon by an electron of velocity v is given by

$$\sigma = \frac{L^3}{v} \sum_{\mathbf{k},s} w_s(\mathbf{k}) = 36\pi e^2 R^2 I_1\left(\frac{\omega R_1}{v}\right) / \hbar v, \qquad (10)$$

where

$$I_n(a) = a^{-1} \int_1^\infty d\xi \, \xi^{-(2n+1)} j_1^2(a\xi) \,. \tag{11}$$

This total cross section consists of the sum of partial cross sections for the creation of plasmons polarized along each of the three Cartesian axes. The cross section $\sigma_{||}$ for creation of a plasmon polarized parallel with the direction of the incident electron is given by

$$\sigma_{||} = (36\pi e^2 R^2/\hbar v) I_2(\omega_1 R/v)$$

while σ_{\perp} , the partial cross section for creation of plasmons polarized perpendicular to the direction of the incident electron, is

$$\sigma_{\perp} = (18\pi e^2 R^2/\hbar v) [I_1(\omega_1 R/v) - I_2(\omega_1 R/v)].$$

In Fig. 2 is shown a plot of the cross sections times $\hbar\omega_1/36\pi e^2 R$ as a function of $\omega_1 R/c$. For values of $\omega_1 R/c$ greater than about 4, the cross section undergoes regular fluctuations of rather small amplitude. These fluctuations have their counterpart in the plane slab case (see Refs. 3 and 4).

IV. RADIATIVE DECAY RATE FOR DIPOLE SURFACE PLASMONS

Now we wish to calculate the transition rate for the process in which a spherical surface plasmon decays by the emission of a photon.

We calculate the interaction Hamiltonian V_{rp} between the plasma and the radiation field from the equation

$$V_{rp} = c^{-1} \int d\mathbf{r} \, \hat{\mathbf{A}} \cdot \hat{\mathbf{J}}_{p}, \qquad (12)$$

where the usual plane-wave expansion is made for the vector potential \mathbf{A} , and the polarization current density \mathbf{J}_p associated with the plasma oscillation is obtained from the perturbation potential $\hat{\boldsymbol{\phi}}$. In the dipole approximation, the matrix element for the transition is

$$\langle \mathbf{p}\lambda \mid V_{rp} \mid s \rangle = (\hbar/i) (\pi R^3 \omega_1^3 / L^3 \omega)^{1/2} (\hat{e}_{p\lambda} \cdot \hat{e}_s), \quad (13)$$

where $\hat{e}_{p\lambda}$ and \hat{e}_s are, respectively, the unit polarization vector of the electromagnetic field and the unit direction vector for the plasma oscillation. **p** and ω are the wave vector and frequency, respectively, of the emitted



FIG. 2. Cross sections for surface plasmon creation by a fast electron. σ_{\parallel} is the cross section for creation of a plasmon polarized parallel with the direction of the incident direction. σ_{\perp} is the cross section for creation of a plasmon polarized perpendicular to the electron direction.

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⁸ P. M. Morse and H. Feshbach, *Methods of Theoretical Physics* (McGraw-Hill Book Co., New York, 1953), Vol. I, p. 1264.

photon. Using the Golden Rule and summing over final photon states, one obtains for the decay rate

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$$\gamma_R = \frac{2}{3} R^3 \omega_1^4 / c^3, \tag{14}$$

where the familiar factor of $2\omega_1^4/3c^3$ reminds one of the average radiation rate of an oscillating electric dipole.

In Sec. V we derive a detailed expression for the distribution of photons following Coulomb-stimulated plasmon decay. In order to show that such photons are likely to be observable experimentally under the proper conditions, we consider the value of γ_R/ω_1 for a 200 Å radius silver sphere. Using a wavelength of ~ 4000 Å = $2\pi c/\omega_1$ for the photon emitted in the dipole plasmon decay, one finds $\gamma_R/\omega_1 = 0.02$ from Eq. (14). If one uses an electronic damping rate γ_D corresponding to the value in bulk silver metal, one may take $\omega_p^2 \gamma_D / \omega^{-3} \sim$ $\epsilon_2(\omega_1) \simeq 0.2$, where $\epsilon_2(\omega_1)$ is the imaginary part of the dielectric permittivity of silver.⁹ Thus $\gamma_R/\gamma_T \sim 0.3$ for this case. In order to obtain the total probability for plasmon excitation followed by radiative decay, one should multiply the probability of excitation by γ_R/γ_T , where γ_R is the radiative decay rate of the plasmon and γ_T is the total decay rate. If one uses Doremus's¹ value of λ_1 for Ag, i.e., about 4000 Å, and considers spheres of radius 200 Å embedded with an average spacing of 2000 Å in a dielectric slab of about 4000 Å thickness, one finds that the total probability for dipole plasmon creation per incident 40-keV electron is $\sim 2 \times 10^{-3}$. Thus the probability of excitation followed by radiative decay is $P_R \simeq 2 \times 10^{-3} \times \gamma_R / \gamma_T \simeq$ 6×10^{-4} . This number should be compared with the probability of exciting normal surface plasmons in a solid Ag foil followed by radiative decay. The radiative decay rate for normal surface plasmons in a solid metal foil is given by

$\gamma_R/\omega_p = (\pi a/\lambda_p) (\sin^2\theta/\cos\theta)$

(see Ref. 4). If one assumes that a = 200 Å and that the angle of observation $\theta = 30^{\circ}$, one finds $\gamma_R/\omega_p \simeq 0.04$. Using $\gamma_T/\omega_p \simeq \epsilon_2(\omega_p) \simeq 0.2$ for plasmons in bulk silver,⁹ one obtains $\gamma_R/\gamma_T \sim 0.2$ for the thin Ag slab. One may use Ferrell's result for the probability of radiative surface plasmon creation by a fast electron normally incident on a thin plasma slab. He finds [Eq. (7b) of Ref. 4] that the maximum probability is 0.0053v/c, which for 40-keV electrons gives $\sim 2 \times 10^{-3}$. Then one finds for this case that the probability of photon emission following Coulomb-stimulated plasma oscillations in a thin silver foil is $P_R \simeq 2 \times 10^{-3} \times \gamma_R/\gamma_T \sim 4 \times 10^{-4}$. Thus emission probabilities are comparable for the two cases considered. Since plasmon decay light from electron bombarded silver foils has been studied in



great detail by many experimenters, it appears that under the proper conditions it should be possible to observe decay photons from spherical plasmons.

V. DIFFERENTIAL CROSS SECTION FOR PHOTON EMISSION BY DIPOLE PLASMONS FOLLOWING EXCITATION BY FAST ELECTRONS

In this section we calculate the differential cross section for the complete process shown in Fig. 3: excitation of a surface plasmon by an electron followed by radiative decay of the plasmon. In this case the transition rate is given by

$$\omega = \frac{2\pi}{\hbar^4} \left| \sum_{\mathbf{k},s} \frac{\langle \mathbf{p}\lambda, \mathbf{k} \mid V_{rp} \mid \mathbf{k}, s \rangle \langle \mathbf{k}, s \mid V_{ep} \mid \mathbf{k}' \rangle}{(\omega_{\mathbf{k}} - \omega_{\mathbf{k}} - \omega_{\mathbf{l}}) + i(\gamma_T/2)} \right|^2 \\ \times \delta(\omega_{\mathbf{k}'} - \omega_{\mathbf{k}} - \omega), \quad (15)$$

where only the time ordering indicated in Fig. 3 has been considered. The ordering in which photon emission precedes plasmon annihilation is not resonant and may be neglected in comparison with the process depicted in Fig. 3 as long as the photon energy $\sim \hbar \omega_1$. In this equation \mathbf{k}' and \mathbf{k} are the wave vectors of the incident and scattered electrons, respectively; \mathbf{p} is the photon wave vector and λ is the polarization index of the photon; s designates the direction \hat{x}_s of the dipole plasmon oscillation, and γ_T is the total plasmon damping rate which is equal to the sum of the radiative damping rate and the electronic damping rate.⁴

One obtains the cross section by summing over the final photon and electron states and dividing by v/L^3 , where v is the velocity of the incident electron, and L^3 is the quantization volume. Finally, if the photon detector responds equally to photons of any polarization, one obtains the following expression for the differential cross section:

$$\frac{d^2\sigma}{d\omega_p d\Omega_p} = \frac{27e^2 R}{2\pi\hbar\omega_1} \left[\frac{\frac{1}{2}\gamma_R}{(\omega - \omega_1)^2 + (\frac{1}{2}\gamma_T)^2} \right]$$
$$\times \left[\sin^2\theta K_2(\omega_1 R/v) + \frac{1}{2}(1 + \cos^2\theta) K_1(\omega_1 R/v) \right], \quad (16)$$

where θ is the angle between the wave vectors of the incident electron and the emitted photon, and ω and ω_1 are the frequencies of the photon and the plasmon. The integrals $K_n(a)$ are equal to $aI_n(a)$, where the I_n are defined in Eq. (11).

⁹ R. H. Huebner, E. T. Arakawa, R. A. MacRae, and R. N. Hamm, J. Opt. Soc. Am. **54**, 1434 (1964).



VI. PHOTON STIMULATION OF DIPOLE PLASMONS

The cross section for plasmon creation by incident photons of frequency ω is given by

$$\sigma_T(2) = \frac{\pi \omega_1 R^3}{c} \frac{\omega_1 \gamma_T}{(\omega - \omega_1)^2 + (\frac{1}{2} \gamma_T)^2}$$

where γ_T is the total damping rate of the dipole plasmon. This result is obtained by employing time-dependent perturbation theory, but allowing for damping of the final state. The probability of dipole plasmon creation by photons of frequency ω incident upon a transparent dielectric slab of dielectric permittivity not greatly different from unity and of thickness t in which the density of spheres is N, is given by $P(\omega) = tN\sigma_T(\omega)$ per photon incident on the slab. One assumes as before that the spheres are randomly spaced with average separation large compared with the wavelength of the photons. Then one may define an absorption coefficient for photons incident upon the system as

$$\chi(\omega) = \frac{\pi N R^3 \omega_1}{c} \frac{\gamma_T \omega_1}{(\omega - \omega_1)^2 + (\frac{1}{2} \gamma_T)^2},$$

which may be compared with the classical absorption coefficient for a dilute system of small, randomly distributed spheres in a medium of dielectric permittivity ϵ^0 , first derived by Mie.¹⁰ In the present notation this may be written

$$\chi_{\rm Mie}(\omega) = \frac{18\pi N\epsilon^0 V}{\lambda_1} \frac{\epsilon_2}{\left[(\epsilon_1 + 2\epsilon^0)^2 + \epsilon_2^2\right]},$$

where $V = \frac{4}{8}\pi R^3$, the volume of each sphere. Here ϵ^0 is assumed constant, while ϵ_1 and ϵ_2 are the real and imaginary parts, respectively, of the frequencydependent dielectric permittivity of the bulk metal of which the spheres are composed. If one sets $\epsilon^0 = 1$, $\epsilon_1 = 1 - (\omega_p/\omega)^2$, $\epsilon_2 = \omega_p^2 \gamma_D/\omega^3$, where γ_D is the bulk electronic damping rate of the metal, and assumes that $\chi_{\text{Mie}}(\omega)$ is strongly peaked about the frequency $\omega_1 = \omega_p/\sqrt{3}$, one finds an expression which agrees exactly with that for $\chi(\omega)$ given above, except that instead of γ_T there appears γ_D , the bulk metal damping rate. The use of $\gamma_T = \gamma_D + \gamma_R$ is clearly a better approximation than γ_D here.

Since much experimental work has been done on the absorption of light in suspensions of small metal spheres, and since the importance of the dipole plasmon state in this work has been amply verified (see, e.g., Refs. 1 and 2), it seems that observation of light emitted in the decay of dipole plasmons should be possible, particularly if the sphere radius is large enough that the radiative damping rate is comparable with the total damping rate. In the example treated in Sec. IV, $\gamma_R/\gamma_T \sim 0.3$ for 200 Å radius silver spheres.

The differential cross section for light emission from a spherical surface plasmon following excitation of the plasmon by light may be determined by the method employed above. The process is shown in Fig. 4. In this case the transition rate is given by

$$w = \frac{2\pi}{\hbar^4} \left| \sum_{s=1}^3 \frac{\langle \mathbf{p} \lambda \mid V_{rp} \mid s \rangle \langle s \mid V_{rp} \mid \mathbf{p}' \lambda' \rangle}{(\omega' - \omega_1)^2 + i(\frac{1}{2}\gamma_T)} \right|^2 \delta(\omega' - \omega),$$
(17)

where \mathbf{p}' and \mathbf{p} are the incident and emitted photon wave vectors, respectively. The nonresonant time order of Fig. 4 has been neglected here.

It is clear that the plasmon created in this process will be linearly polarized in the same direction as the polarization vector of the incident photon. Summing over final photon states, we obtain the following differential cross section:

$$\frac{d\sigma}{d\Omega_{\rm p}} = \frac{3R^3\omega_1^2}{4c} \frac{\frac{1}{2}\gamma_R}{(\omega - \omega_1)^2 + (\frac{1}{2}\gamma_T)^2} \, \frac{1}{2} [1 + \cos^2\theta], \quad (18)$$

where θ is the angle between the wave vectors of the incident and emitted photons, ω is the frequency of the emitted photon, R is the radius of the sphere, and γ_R is the radiative decay rate given in Eq. (14).

VII. SUMMARY

We have considered the interaction of fast but nonrelativistic electrons with dipole plasmons in spherical systems. We find that the yield of photons from the decay of dipole plasmons which have been generated by fast electron bombardment of fine spheres suspended in a transparent medium may be comparable with the corresponding yield from thin metal slabs. It appears that experimental work along these lines would be quite interesting. Further work planned in this area includes the study of plasmons of higher order, as well as volume plasmons, in spherical systems.

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¹⁰ G. Mie, Ann. Physik (4) **25**, 377 (1908); also, M. Born and E. Wolf, *Principles of Optics* (Pergamon Press, Inc., New York, 1959), p. 630.