Bulk-Plasmon Dispersion Spectrum of Be Using an X-Ray Scattering Technique*

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Spectroscopic analysis of scattered x rays from Be irradiated by copper-target $K\beta$ radiation establishes the existence of a first-order bulk-plasmon line. Dispersion of the plasmon energy as a function of the scattering angle for momentum transfer $\hbar\vec{k}$ smaller than the critical $\hbar\vec{k}_c$ was observed, as predicted by Bohm and Pines, and Nozières and Pines. The plasmon energy for scattering angle zero was found to be 19.4 ± 0.4 eV. No appreciable dispersion was observed for $\hbar\vec{k} > \hbar\vec{k}_c$. Adopting the Drude-Zener model for the electron gas, the experimental half-width of the unfolded lines gives an estimated plasmon lifetime of the order of 1.4×10^{-16} sec, for scattering angle $\phi = 0$.

I. INTRODUCTION

It has been found[1] that plasma oscillation can be excited in Li, C, and Be through x-ray scattering. The characteristic Cr $K\beta$ x-ray line, scattered at an angle $\phi = 15°$ from these elements, was found to undergo a distinct energy loss equal to the plasmon energy of the corresponding elements. Extending the measurements to 10° and 5°, it was reported initially that the plasmon energy was independent of the scattering angle. However, a detailed examination[2] showed a slight shift of the peak of the plasmon line to longer wavelengths. This has been attributed to the dispersion of plasmon energy as a function of the momentum transfer. Similar results have been reported recently.[3]

Adopting a simplified description of a conductor, whose elements are (i) a uniform background of positive charge which replaces the positive ions and (ii) the interacting electron gas, Bohm and Pines[4] examined the response of such a system, the so-called quantum plasma, to a suitable external perturbation. This perturbation creates a local modification in the state of the interacting electron gas (initially in equilibrium with the positive background) resulting in a tendency of the electrons to reestablish electrical neutrality. The movement of the ions is negligible, while a definite momentum is transferred to the plasma. For adequately small momentum transfer $\hbar\vec{q}$, an oscillatory motion of the electrons appears, corresponding to periodic variations of the charge density. This is a longitudinal oscillation with an angular frequency ω_p, given by the fundamental relation

$$\omega_p = (4\pi n e^2/m)^{1/2} . \tag{1}$$

The relation (1) is derived under the assumption that the influence of the thermal agitation of free electrons is negligible.[5] However, the thermal agitation of the individual electrons is opposed to the coherent plasma oscillations. In the case of an oscillation propagating with a wave vector $\vec{k}$, the thermal agitation introduces a corrective term to the relation (1). A reasonable measure of the average electronic thermal energy is of the order $\hbar\langle (kv_i)\rangle_{av}$ which for valence electrons in metals can be estimated as $\hbar k v_0$, where v_0 is the Fermi velocity. This individual agitation gives rise to a dispersion relation of the approximate form

$$\omega_k^2 = \omega_p^2 + k^2 v_0^2 . \tag{2}$$

If $\omega_p^2 \gg k^2 v_0^2$, the collective character of the excitation dominates, whereas in the absence of this condition, single-particle excitations assume a significant role. The transition between these two domains occurs at a critical value of k which is given by

$$k_c = \omega_p / v_0 . \tag{2'}$$

When an x-ray or electron beam disturbs a quantum plasma, its momentum changes by an amount taken up by a collective oscillation of the plasma.[1,6] If the incident photon has a wave vector $\vec{k}_1$ and the scattering angle is ϕ, the change of momentum $\hbar\vec{q}$ is given by the relation

$$\hbar\vec{q} = 2\hbar\vec{k}_1 \sin\tfrac{1}{2}\phi \tag{3}$$

This $\hbar\vec{q}$ is exactly the momentum transferred to the plasma, and hence equal to the momentum $\hbar\vec{k}$. Thus in scattering experiments, the plasma excitation will dominate over any kind of individual excitation for angles smaller than ϕ_c given by

$$k_c = 2k_1 \sin\tfrac{1}{2}\phi_c \tag{3'}$$

In this scattering event, an energy equal to the plasmon energy $\hbar\omega_k$ is given to the plasma and thus the initial energy of the scattered photon is decreased by an equal amount.

The dispersion relation (2) actually represents only a qualitative approach to the subject. According to the dielectric treatment of the electron gas[7] in the random phase approximation (RPA), the true dispersion relation should, to a good degree of approximation, take the form

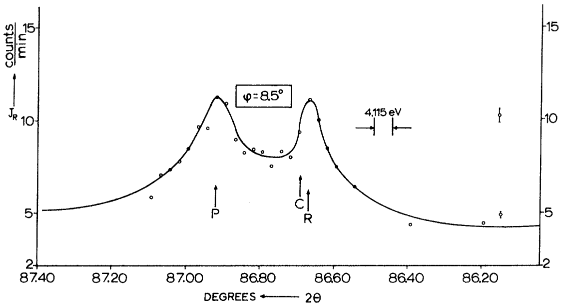

FIG. 1. Spectrum of the radiation after scattering on Be under scattering angle $\phi = 8.5°$; R, primary line; C, position of Compton peak; P, plasmon line.

$$\hbar\omega_k = \hbar\omega_p + \tfrac{12}{10}(\hbar^2 v_0^2/\hbar\omega_p)k_1^2 \sin^2(\tfrac{1}{2}\phi). \quad (4)$$

The energy-loss spectrum calculated in Ref. 7 is found to contain not only a component with energy shift equal to $\hbar\omega_k$, but also other components due to individual electron excitations, corresponding to intra- and interband transitions. The interband transitions produce a damping of the plasma oscillation, which results in a finite width of the plasma line. Besides the individual excitations, the lifetime of the plasma oscillations is reduced by the emission of electromagnetic radiation. Thus, the total lifetime will be given by

$$1/\tau_{tot} = 1/\tau_{exc} + 1/\tau_{rad} .$$

For zero momentum transfer ($\phi \to 0$), no radiation is emitted; hence

$$\tau_{tot} = \tau_{exc} .$$

According to the Drude-Zener approximation for the electron gas, we obtain

$$\tau_{tot} = \tau_{exc} = \hbar/\gamma_0 , \quad (5)$$

where γ_0 is the corresponding half-width in electron volts for $\phi = 0$. From relation (5) we find a lifetime of the plasmon which is due only to the interband excitations.

In the present work, we have repeated the former[1, 2] experiment on Be using x rays of a different wavelength and a different apparatus. Our measurements were extended to higher scattering angles. Statistical accuracy was also increased.

II. EXPERIMENT AND RESULTS

A copper x-ray tube (25 mA, 40 kV, both stabilized) was used to irradiate, in a transmission position, a slab of polycrystalline Be held at a distance of 12 cm from the source. A lead cylinder located at the exit of x-ray tube reduced the divergence of the scattering angle to ±1.8 °.

The scattered radiation entered a Soller system (length 25 cm, 100 channels of 100 μ width) and was analyzed on a Philips diffractometer with a $(22\bar{4}3)$ quartz crystal. Both the scatterer and the analyzing crystal were held in evacuated chambers with Mylar windows.

The spectrum was measured point by point with a Geiger counter at angles 2Θ differing by 0.025 °. The scattering experiments were carried out with the $K\beta$ line ($\lambda = 1.392$ Å). The $K\beta$, although less intense than $K\alpha_1$ and $K\alpha_2$, is very convenient as a primary line because of the absence of other lines in its vicinity. We thus obtained a single plasmon line without any overlapping. The spectral distribution of the intensity scattered by Be has been measured for 11 different scattering angles ϕ, i.e., 6.5°, 8.5°, 10.2°, 11.0°, 14.0°, 17.0°, 18.5°, 20.0°, 22.5°, 25.0°, 44.5°. By this choice of angles we avoided the Debye-Sherrer lines, as far as possible. The spectral distribution for $\phi = 8.5°$ is shown in Fig. 1. It consists of three components indicated in the figure by the arrows R, C, and P. The feature labelled R is due to the thermal diffuse scattering (TDS) and coincides with the primary line. The mark at C indicates the position of the maximum of the Compton band as calculated for a free electron at rest, while that at P represents the plasmon oscillation line. In Fig. 2 we have plotted the spectrum for $\phi = 6.5°$, 10.2°, and 14.0°. We observe clearly a slight shift of the plasmon line. In Fig. 3 we have plotted the spectrum for $\phi = 17.0°$, 20.0°, 25.0°, and 44.5°. In the first three cases, although the momentun transfer is higher than the critical, a band of the same shape and at almost the same energy continues to be observed, but practically without dispersion. As we explain below, this line does not fulfill the requirements for individual electron excitations according to the Glick-Ferrell theory.[8] At $\phi = 44.5°$, the plasmon line is no more observable; it might, however, contribute in some way to the background. The high intensity of the unshifted radiation is partially due to coherently scattered primary radiation as the scattering angle of 44.5°

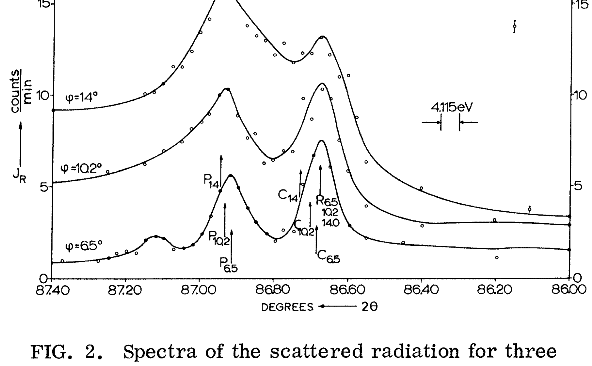

FIG. 2. Spectra of the scattered radiation for three different scattering angles $\phi < \phi_c$.

is very near to the value $\phi = 44.7°$ of the {020} line. To be sure that the observed spectrum is due to the radiation scattered on Be, we performed additional experiments taking the spectrum without any scatterer in position. The spectra obtained are composed of a uniform background with intensity of around 16 counts/min. For scattering angle of 6.5°, we observe, in addition to the uniform background, an R band. This is radiation passing through the Soller after being scattered on its front face for geometrical reasons. In Figs. 1–3, the intensities plotted are those remaining after subtracting the intensities found without a scatterer in position.

As mentioned above, we observe a shift of the plasmon line upon increasing the scattering angle ϕ. Under the present circumstances, ϕ_c is calculated to be 16° after setting $v_0 = 2.25\times 10^8$ cm/sec and $\hbar\omega_p = 19$ eV in formulas (2′) and (3′).

For a better treatment of the present results, we classify them in two cases, according to whether the momentum transfer is smaller or larger than the critical one.

Taking the energy difference between the maximum of primary line and the maximum of plasmon line for various scattering angles, we find the plasmon energy to be a function of the momentum transfer to the system. It is plotted as a function of q^2 in Fig. 4. On this graph we observe that for small angles ϕ, up to approximately 11°, the dispersion follows very closely a linear relation in accordance with Eq. (4), i.e., the shift of plasmon energy is proportional to the square of the momentum transfer. Extrapolating towards zero scattering angle we find a plasmon energy $\hbar\omega_p = 19.4 \pm 0.4$ eV. The slope of this line is

$$2.62\times 10^2 \text{ eV} = 0.65\times 10^2 \text{ eV/rad}^2 \ .$$

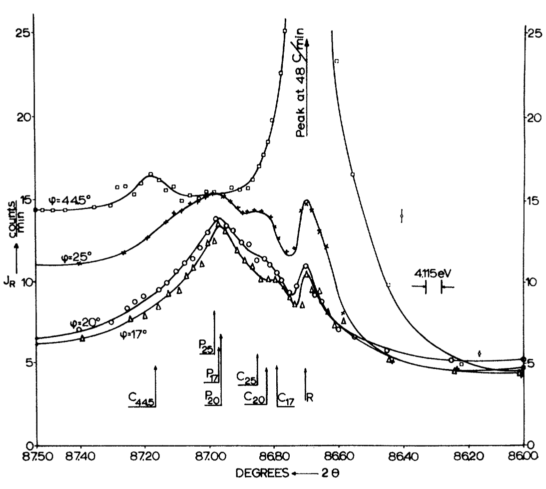

FIG. 3. Spectra of the scattered radiation for four different scattering angles $\phi > \phi_c$.

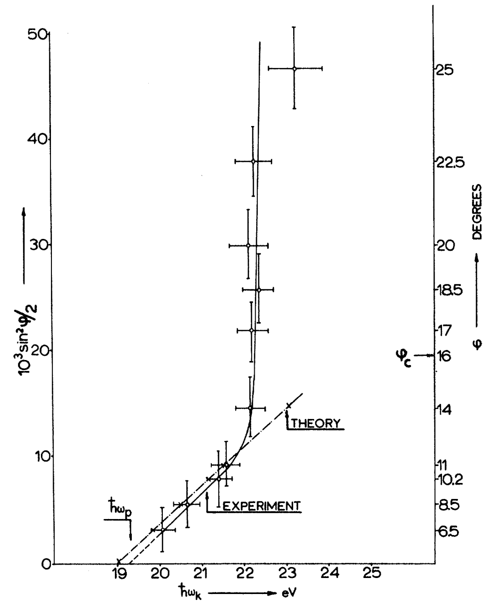

FIG. 4. Experimental dispersion curve for the bulk plasmon of Be.

For comparison, we plot the straight line resulting from free-electron theory according to Eq. (4) in which we insert $\hbar\omega_p = 19$ eV as calculated from (1) assuming 2 electrons/atom and $v_0 = 2.25\times 10^8$ cm/sec, yielding a slope equal to

$$2.8\times 10^2 \text{ eV} = 0.71\times 10^2 \text{ eV/rad}^2 \ .$$

The resulting mean value of plasmon energy, 19.4 eV, is higher than the theoretical free-electron value of 19 eV. Higher values have also been found by other workers,[1,3,6] using various methods of measurement. This can be attributed to a small contribution of the bound electrons to the collective oscillation.

At angles near 14° we notice that $\hbar\omega_k$ is smaller than anticipated. This might be attributed to an error in the determination of the position of the plasmon peak in Fig. 2, caused by the increased contribution of the Compton band which approaches the plasmon line as scattering angles increase. Beyond the critical momentum transfer the dispersion almost ceases; the plasmon line is still observable, although in this region the RPA does not advocate in favor of plasma oscillation. Bohm[8] points out that plasmons may be produced at such momentum transfers, but the dominating effect in the loss spectrum will be that of the individual electron excitations. We could claim that even in these cases plasma oscillations are excited, but with shorter lifetime. Presumably, it can not be attributed to interband transitions because Glick and Ferrell[9] have suggested that the single-electron excitation spectrum exhibits a remarkable disper-

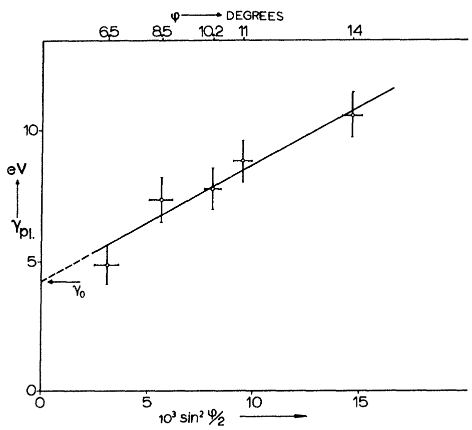

FIG. 5. The real half-width of the plasmon line as a function of q^2.

sion as the momentum transfer increases, starting just beyond the plasmon cutoff (momentum $\hbar q_c$). Also, the peak of this spectrum is located at twice the frequency observed in the present experiment. Nevertheless, just beyond the plasmon cutoff we observe a sharp decrease in the slope of the dispersion curve, which indicates a change in the phase of the interacting system.

We will now discuss the plasmon lifetime. As mentioned in Sec. I, the individual excitations introduce a damping even for $\vec{q}$ approaching zero. The plasmon lifetime can be determined from the half-width of the plasmon line. The measured γ_{expt} half-width is larger than the real one because the primary radiation had a finite width b; because of the folding of the plasmon line with the primary line, the real width can be obtained, with a good approximation, from the relation

$$\gamma_{pl}^2 = \gamma_{expt}^2 - b^2 , \tag{6}$$

where γ_{pl} is the real half-width of the plasmon line.

In Fig. 5 we plot the values of γ_{pl} thus determined. From this, we obtain γ_0 which when inserted in (5) yields

$$\tau_{exc} = 1.4 \times 10^{-16} \text{ sec.}$$

For scattering angles above ϕ_c, the half-width could not be determined with any accuracy because of excessive peak broadening.

The height of the plasmon peak as a function of scattering angle is plotted in Fig. 6.

III. CONCLUSION AND DISCUSSION

We will now try to prove the claim that the so-called plasmon component is due to an energy shift associated with collective oscillations.

The experimentally determined shift of 19.4 eV found in the present experiment is close to the 21.5 eV found in similar experiments, using longer wavelengths[1,3]; however, the difference seems to be slightly beyond the experimental error. It also coincides with the region of values found from characteristic energy losses of electrons.[6,10] By applying free-electron theory for bulk plasmons,[11,12] one also arrives at approximately the same value. A further argument in favor of our claim is the fact that Fig. 4 shows a dispersion that follows the relation (4) very well as long as the momentum transfers are smaller than the critical.

The half-width for an ideal crystal, at scattering angles near zero ($\gamma_{rad} \rightarrow 0$), is likely to be identical with that of the primary line, satisfying the condition that the spectrum of the plasmon scattering is approximately of the δ-function type, as predicted by RPA theory. The larger width found in the present experiment (Fig. 5) might be attributed to the scattering of a plasmon on the space-varying structure[13] of our polycrystalline sample. As the scattering angles increase, we find that the width increases proportionally to q^2 as predicted by the theory of DuBois and others.[14–16] An attempt to explain the newly observed[17] line as a Compton RPA peak, adopting the Ohmura and Matsudaira[18] model, is not fruitful for the following reason: Beryllium has an electron density that corresponds to $r_s = 1.9$. With a good approximation we can use Eq. (4.2) or Fig. 3 of Ref. 18 to find the position of the RPA peak. The example calculated therein refers to $\phi = 15°$ and $\lambda = 1$ Å from which a momentum transfer of 1.64×10^8 cm^{-1} can be calculated. The corresponding peak is shifted by 29.5 eV. In the present experiment, the momentum transfer mentioned corresponds to $\phi = 20°$. Hence, in our curve for $\phi = 20°$, the RPA peak of the Compton profile should occur at 29.5 eV, and in no way can be caused by the peak which, as seen in Fig. 3, occurs at a shift of about 22 eV.

In recent measurements[17] carried out by Schülke

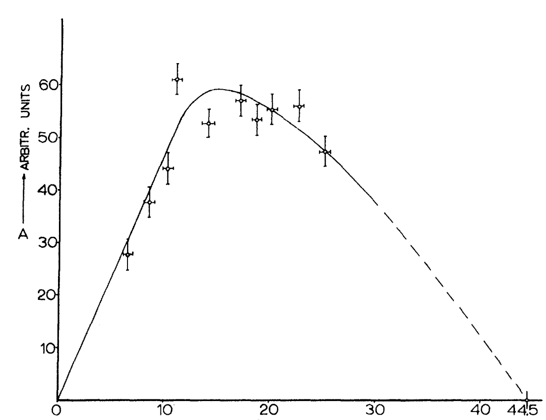

FIG. 6. The height of the plasmon peak as a function of scattering angle.

et al., the existence of a plasmon line was not reported, even for small values of the momentum transfer. This, we assume, is due to their very large experimental width which is of the order of 22 eV and therefore would overlap the plasmon band. In our experiments the experimental width is of the order of 11 eV.

In conclusion, we observe that in Fig. 2 the peak of the plasmon line is well separated from TDS and Compton lines which overlap. In Fig. 3 the spectra for $\phi = 17°$, $20°$, and $25°$ allow the separate observation of the TDS and the plasmon peak. The Compton band is observed as a weak bump.

The observed line is due to the bulk plasmon and no attempt has been made to detect higher-order plasmons. However, in the case of $\phi = 6.5°$ it might be possible to attribute the bump at the far left to a second-order plasmon.

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Channeling of Positrons

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Axial and planar channeling in thin single-crystalline gold foils has been investigated by wide-angle scattering of monoenergetic positrons. The beam was obtained by accelerating the positrons emitted from a Co^{58} source in a 1-MeV Van de Graaff. The results are in good agreement with corresponding measurements for protons. For the planar case, classical calculations are compared to calculations based on the dynamical theory of diffraction. The results are very similar except for the "wiggles" due to wave interference, which appear in the quantum-mechanical calculation. These, however, are difficult to resolve experimentally.

INTRODUCTION

The aim of this experiment is to shed some light on the question of applicability of classical channeling theory to the directional effects observed for emission of electrons and positrons from a single crystal. In Uggerhøj's measurements[1,2] of the angular distribution of electrons and positrons emitted from Cu^{64} implanted in copper single crystals, a quantitative comparison with theory or with heavy-particle channeling was difficult because of the radiation damage incurred during the implantation of the radioactive ions into the crystal. The results for positrons, however, were consistent