frequencies the temperature difference would be higher.

One might believe that strain amplitude could be the cause of this difference in the positions of the two lines, and it might be the case at 40 kc/sec. At 5 Mc/sec, however, this is not likely and it is still more unlikely that the two together would produce this result.

One factor that does seem to be consistent is the purity of the samples. The aluminum used both by Hutchison and Filmer and by Bordoni is said to have a purity of 99.994%. The purity of the aluminum used in our experiments was nominally 99.80% which of course is very different from the higher purity.

In order to try to settle this question of the effect of purity, experiments on high-purity aluminum are now being undertaken in our laboratory both on single crystals and on polycrystalline aluminum.

Rough estimates of the ratio of the Peierls stress τ_P^0 to the shear modulus G based on the megacycle data yield $\tau_P^0/G=1.2\times10^{-5}$. Similar rough estimates of the width of the dislocation kink (based on reference 3) yield $w=170a$, where a is the lattice constant.

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Characteristic Energy Loss of Electrons in Graphite[*]

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The problem of analyzing the characteristic energy loss of electrons in solids is discussed in some detail. Based on the general discussion developed in Secs. 1 and 2, the observed 7.5-ev loss line of graphite is tentatively identified as due to the collective excitation of π electrons. The theoretical analysis gives results which strongly favor this identification. Furthermore, it has been shown that the present theory together with detailed experimental studies is very useful for the determination of the π-electron band structure of graphite.

I. INTRODUCTION AND SUMMARY

IN recent years,[1] many experiments have been carried out to observe the energy spectrum of electrons passing through thin solid foils, of thickness of the order of several hundred angstroms. Improvements in the experimental techniques made it possible to get an energy resolution of about 1 ev. The observed energy spectrum consists of several fairly sharp lines, whose positions are independent of the incident energy. The number of lines seems to increase with the thickness of the foils. The angular dependence of the energy loss has also been observed[2,3] as well as the angular dependence of the intensity,[4] though the results obtained by different workers are not always in agreement with each other.

The essential facts, which are not favorable to the simple interpretation of the results as a single-electron excitation due to the interband transition, are the sharpness of the line width and the strong angular dependence of the observed energy loss. Although the interpretation of the observed energy loss is not conclusive yet, the proposed identification of the transition involved in the formation of the discrete spectra may be classified as follows:

(A) single-electron excitation of atomic origin,
(B) single-electron excitation of crystalline origin,
(C) collective excitation of valence electrons.

An example of type (A) is a transition in the soft x-ray region.[5] A well-known process of type (B) is the transition from a localized, bound state to one of the energy bands above the Fermi level, which is believed to be responsible for the fine structure of the K or L x-ray absorption edge.[6,7] Transitions of type (C) were first suggested by Pines on the basis of the plasma theory.[8]

To utilize the energy-loss experiments as a tool for exploring the structure of matter, we must carefully set up criteria which enable us to identify the loss lines with specified transitions. First of all, if an observed line is of atomic origin, the corresponding line must be observed in the gaseous state of the element, as well as in some chemical compounds, probably with a slight and systematic shift of position. If a loss line, however, is observed only in the crystalline state of the element, it may be due either to a single-electron transition of

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[1] Marton, Leder, and Mendolowitz, Advances in Electronics 7, 185 (1953); L. Marton, Revs. Modern Phys. 28, 172 (1956).

[2] F. Leonhard, Z. Naturforsch. 69, 727, 1019 (1954).

[3] H. Watanabe, J. Phys. Soc. Japan 11, 112 (1956).

[4] Marton, Simpson, and McGraw, Phys. Rev. 99, 495 (1955).

[5] L. Marton and B. Leder, Phys. Rev. 94, 203 (1954).

[6] Leder, Mendolowitz, and Marton, Phys. Rev. 101, 1460 (1954).

[7] R. L. Kronig, Z. Physik 70, 317 (1931); 75, 191, 468 (1932); T. Hayashi, Sci. Repts. Tohoku Univ. 33, 123,183 (1949); 34, 185 (1950).

[8] D. Pines, Revs. Modern Phys. 28, 184 (1956).

type (B), or to the collective excitation of the valence electrons. It is rather difficult to discriminate between these two alternatives. The most conclusive discrimination would be obtained by measuring the angular dependence of the energy loss, though for some cases this measurement also appears to be not conclusive. In Sec. 2, we shall discuss this point in detail. Ferrell has pointed out that the angular dependence of the intensity should not be relied upon to distinguish between the two alternative identifications (B) and (C).[9]

Crucial tests for discriminating between (B) and (C) may be carried out in the following ways[10]:

(I): The dependence of the intensity of multiple loss peaks on thickness can depend only on the mean free path for the discrete excitation under consideration in case (B), while it may also be influenced by the excitation of higher harmonics in the collective excitation (C).[11] This influence becomes important especially as the thickness of a foil decreases. In many cases, however, the presence of a single-electron excitation line near the double-energy position of the line under consideration may disturb the application of this criterion.

(II): The fine structure of the K or L absorption edge seems to correlate with the discrete energy loss of an electron. Since x-ray absorption is a one-quantum process, the correlation with the loss line due to multiple excitations by an electron should decline to a great extent, because such an x-ray absorption can take place only for a higher order process which is very unlikely to occur. Therefore, the existence of a single-electron excitation line, which could be confused with a multiple excitation line, can be examined by comparison with the x-ray absorption line. The combination of these two methods may serve our purpose.

In Sec. 3, we shall discuss the collective excitation of π electrons in graphite. The observed 7.5-ev loss line is identified as due to the collective excitation of π electrons, based on the discussion developed in Sec. II. From the observed value 7.5 ev, we can determine a resonance energy γ_1 of the nearest neighbors *between* planes. The resonance energy γ_0 of the nearest neighbors *in* the plane is responsible for the dependence of $\hbar\omega_p$ on the angle, Θ, between the direction of the incident electron beam and the c axis of the graphite crystal. Various features related to the Θ dependence of $\hbar\omega_p$ will be discussed in detail. The dependence of the energy loss on the deflection angle θ is also examined, and it is shown that the calculated coefficient of the θ^2 term is in good agreement with the observed result.

Though the present investigation is concerned with the special case of graphite, it shows that the electron energy-loss experiments provide very useful tools for exploring the electron structure of solids, if one does the experiments with a single crystalline thin foil. Furthermore, it can be seen after the discussions of Sec. II and III that the Θ dependence of the energy loss is a characteristic feature of the plasma excitation. If the energy loss is due to the single-electron excitation, process (B), it does not show such a Θ dependence. Therefore, the observations of the Θ dependence of the energy loss, together with that of the θ dependence, furnishes a definite criterion which discriminates between the processes (B) and (C).

It may be interesting to develop systematic studies of the energy loss of various organic compounds which have conjugated double bonds, because they may exhibit a characteristic behavior of π electrons of the conjugated double bonds.[12]

II. ANGULAR DEPENDENCE OF ENERGY LOSS

Watanabe[3] has observed the angular dependence of the energy loss $\Delta E(\theta)$, and determined parameters α and β, by expressing $\Delta E(\theta)$ as

$$\Delta E(\theta)=\Delta E(0)+\alpha E\theta^2+\beta E\theta^4, \tag{1}$$

where E is the kinetic energy of the incident electron. The values determined for α and β are given in Table I. The table contains also the energy loss at $\theta=0$ and the calculated values $\hbar\omega_p, \alpha_B$ from the Bohm-Pines dispersion relation,

$$\hbar\omega=\hbar\omega_p+\alpha_B E\theta^2+\beta_B E\theta^4, \tag{2}$$

where ω_p is defined by the well-known formula

$$\omega_p{}^2=(4\pi e^2/m)n, \quad n=\text{density of electrons} \tag{3}$$

and α_β and β_B are given as

$$\alpha_B=\frac{2}{nV}\sum_p \frac{p_2}{2m}\Big/\hbar\omega_p, \quad \beta_B=E/2\hbar\omega_p. \tag{4}$$

Comparing α with α_B, Watanabe concluded that the sharp lines observed in Be, Mg, Al were undoubtedly

TABLE I. Observed data[3] of the energy loss ΔE and the angular-dependence coefficients α and β. Calculated values of the plasmon excitation energy and α, based on the Bohm and Pines theory are also exhibited in the third and fifth columns.

Material	ΔE ev	$\hbar\omega_p$ ev	α	α_B	$\beta\times10^{-2}$
Be	19	19	0.84 ± 0.08	0.9	···
Mg	10.5	11	1.24 ± 0.08	0.88	4 ± 1
Al	15	16	1.00 ± 0.10	0.9	3 ± 1
Ge	16.5	15.5	1.66 ± 0.30	0.9	···
Graphite	7.5	12.3[a]	2.0 ± 0.6	0.8[a]	···

[a] Calculated value, by assuming that π electrons are free.

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[9] R. A. Ferrell, Phys. Rev. **101**, 554 (1956).

[10] The discussions presented here, as well as in the next paragraph, are due mainly to Professor Hayakawa. The author wishes to express deep thanks to Professor Hayakawa for his helpful discussions.

[11] Blackstock, Richica, and Birkhoff, Phys. Rev. **100**, 1078 (1956). The rather systematic discrepancy between the experimentally determined mean free path and the calculated mean free path of Bohm-Pines theory may be regarded as an evidence of the multiple plasmon emission.

[12] G. Araki and T. Murai, Progr. Theoret. Phys. Japan **8**, 639 (1952).

caused by the excitation of collective oscillations, since the angular-dependence parameters α were in agreement with the corresponding parameters α_B obtained from the Bohm-Pines dispersion relation.

However, it should be pointed out that such an angular dependence of the energy loss is not necessarily considered as a characteristic feature of the Bohm-Pines dispersion relation. To show that, let us consider the single-electron excitation between an occupied band and an excited band. Then, the energy-momentum conservation is

$$\frac{p_0^2}{2m}+E_n(\mathbf{q}_0)=\frac{p^2}{2m}+E_m(\mathbf{q}), \tag{5}$$

$$\mathbf{p}_0+\mathbf{q}_0=\mathbf{p}+\mathbf{q}, \tag{6}$$

where $\mathbf{p}_0$, $\mathbf{q}_0$ are the momenta of the incident and the target electrons, $\mathbf{p}$, $\mathbf{q}$ are those of the scattered and the excited electrons, respectively. Since we are interested in the case of small momentum transfer, we are not considering the umklapp processes. From these equations, we get the energy loss of an incident electron, $\Delta E(\mathbf{q}_0)$ as

$$\Delta E(\mathbf{q}_0)=\frac{1}{2m}(p_0^2-p^2)=E_m(\mathbf{q}_0+\Delta\mathbf{p})-E_n(\mathbf{q}_0), \tag{7}$$

where $\Delta\mathbf{p}=\mathbf{p}_0-\mathbf{p}$ is the momentum transfer of the incident electron. For the limit of small $\Delta\mathbf{p}$, we may expand $E_m(\mathbf{q}_0+\Delta\mathbf{p}_p)$ as

$$E_m(\mathbf{q}_0+\Delta\mathbf{p})=E_m(\mathbf{q}_0)+(\Delta\mathbf{p}\cdot\nabla)E_m(\mathbf{q}_0)+\tfrac{1}{2}(\Delta\mathbf{p}\cdot\nabla)^2E_m(\mathbf{q}_0). \tag{8}$$

Thus, $\Delta E(\mathbf{q}_0)$ is written as

$$\Delta E(\mathbf{q}_0)=\{E_m(\mathbf{q}_0)-E_n(\mathbf{q}_0)\}+(\Delta\mathbf{p}\cdot\nabla)E_m(\mathbf{q}_0)+\tfrac{1}{2}(\Delta\mathbf{p}\cdot\nabla)^2E_m(\mathbf{q}_0). \tag{9}$$

If there is no external field which may give a preferred direction in the crystal, because of a reflection symmetry of the band structure, the contributions of the second term with positive values are balanced off by those with negative values. For small deflection angles θ, $\Delta\mathbf{p}$ can be written as

$$\Delta\mathbf{p}=p_0\theta\mathbf{e}, \tag{10}$$

where $\mathbf{e}$ is a unit vector which is perpendicular to the direction of incident momentum $\mathbf{p}_0$. Hence, we get

$$\Delta E(\mathbf{q}_0)=\{E_m(\mathbf{q}_0)-E_n(\mathbf{q}_0)\}+m(\mathbf{e}\nabla)^2E_m(\mathbf{q}_0)\cdot E\theta^2, \qquad (11)^{13}$$

where $E=p_0^2/2m$. Here, the average of the coefficient of $E\theta^2$, corresponding to the case of random orientation of the single crystal or the case of a polycrystal, may nearly be equal to unity for very highly excited bands, though generally it may take positive values as well as negative values, depending on the electronic state. The angular independent term exhibits the width of the initial momentum distribution of the target electrons, which would be of the order of several ev. Thus, the above discussion gives us a conclusive criterion for excluding the possible interpretation of the energy loss as due to a single-electron band-band transition.

Our interpretation of the 7.5-ev loss line of graphite as due to the collective excitation of π electron is, on one hand, based on the experimental fact that the 7.5-ev line is lacking for some carbon compounds. On the other hand, it is supported by our theory which gives the collective eigenfrequency corresponding to 7.5 ev as well as an angular dependence parameter α of 2.

III. COLLECTIVE EXCITATION OF π ELECTRONS IN GRAPHITE

Though we have some evidence that the 7.5-ev loss line in graphite may be due to the excitation of collective oscillations of π electrons, a remarkable discrepancy between the observed 7.5-ev loss and the free π-electron plasma frequency has not been understood yet. Here, based on the general theory of the collective oscillation,[14] we shall analyze in detail the experimental results for graphite.

Since, at absolute zero temperature, the first zone of electron state in graphite is completely filled up, the frequency of collective oscillation of the π electron is given by

$$\omega_p{}^2=\frac{4\pi e^2}{m}\frac{1}{V}\sum_{\mathbf{p}\cdot\lambda}\left\{1-\frac{m}{\hbar^2}(\mathbf{e}\nabla_p)^2E_n(\mathbf{p})\right\}N(p\cdot\lambda), \tag{12}$$

where $\mathbf{e}$ is a direction vector of momentum transfer. It is perpendicular to a direction of momentum of incident electron for small deflection angle θ. In the following discussion, we will assume the electron beam incident on a single-crystal foil at angle Θ with the c axis.

Now, Wallace's theory[15] gives the following expression for the energy band,

$$E(\mathbf{p})=E_0-\gamma_1\cos(cp_x/2)-[\gamma_1^2\cos^2(cp_x/2)+\gamma_0^2|S|^2]^{\frac{1}{2}}, \tag{13}$$

where γ_0 is the resonance energy between the nearest neighbors *in* the plane, γ_1, is that of the nearest neighbors between planes. S is defined as

$$S=\exp[-ip_x(a/\sqrt{3})]+2\cos(p_ya/2)\exp[ip_x(a/2\sqrt{3})], \tag{14}$$

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[13] Contrary to Eq. (11), for the in-band single-electron excitation in the unfilled valence band, the average excitation energy can be calculated by the Fermi gas model and it gives a linear independence on θ as follows,[9]

$$\frac{4E_0}{3E}\frac{\theta}{\theta_0}\left[1-\frac{1}{12}\left(\frac{\theta E_0}{\theta_0 E}\right)^2\right]^{-1},$$

where E_0 is the Fermi energy and $\theta_0E=p_0/p$. The author would like to thank Dr. R. A. Ferrell for his kindness in calling the author's attention to this point.

[14] Y. H. Ichikawa, Progr. Theoret. Phys. Japan **18**, 247 (1957).

[15] P. R. Wallace, Phys. Rev. **71**, 622 (1947).

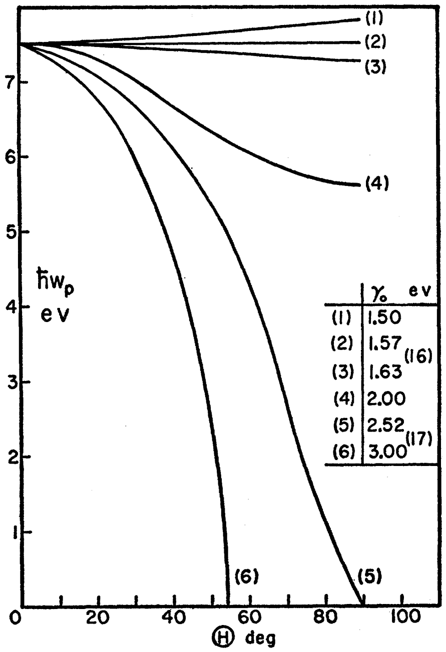

FIG. 1. The Θ dependence of the π-electron plasma frequency in graphite.

where $a=2.46$ A, $c=7.72$ A are the magnitudes of the lattice translational vectors, respectively.

Usually, people transform the volume integral $\sum_{\mathbf{p}\cdot\lambda}$ into a surface integral by using a relation

$$\sum_{\mathbf{p}\cdot\lambda}\cdots=\frac{2V}{8\pi^3}\int d^3p\cdots$$
$$=\frac{V}{4\pi^3}\int d\epsilon\int dp_x dp_y\frac{1}{(\partial\epsilon/\partial p_z)_{\text{Surface}}}. \quad (15)$$

However, the above transformation is a very undesirable procedure to carry out, since it is well known that Wallace's theory is inaccurate near the surface of a hexagonal Brillouin zone and thus it may introduce serious errors due to the defect of Wallace's theory. So, instead of that, we will approximate $E(\mathbf{p})$ by an expansion at the center of Brillouin zone and carry out the volume integral. Errors introduced by such effective-mass approximation would be less serious than one might suspect, because there occurs no cancellation between terms in second order of $\mathbf{p}$.

Finally, we get

$$\langle(\mathbf{e}\cdot\nabla)^2E(\mathbf{p})\rangle=\tfrac{1}{2}\gamma_0a^2\sin^2\Theta+\tfrac{1}{4}\gamma_1c^2\cos^2\Theta, \quad (16)$$

where the average is taken over the orientation of the crystal in the $x-y$ plane. Thus, the ω_p^2 becomes

$$\omega_p^2=\frac{4\pi e^2}{m}n\left[1-\frac{m}{m(\Theta)^*}\right], \quad (17)$$

where

$$\frac{1}{m(\Theta)^*}=\frac{1}{2\hbar^2}\left\{\sin^2\Theta+\frac{1}{2}\frac{\gamma_1}{\gamma_0}\left(\frac{c}{a}\right)^2\cos^2\Theta\right\}a^2\gamma_0. \quad (18)$$

From this result, we can draw the following results:

(1) ω_p^2 for $\Theta=0$ depends only on γ_1 as

$$\omega_p^2(0)=\frac{4\pi e^2}{m}n\left[1-\frac{m}{4\hbar^2}c^2\gamma_1\right]. \quad (19)$$

To give $\hbar\omega_p=7.5$ ev, which is obtained by Watanabe using a thin foil of single crystal, γ_1 must be chosen to be

$$\gamma_1=0.32 \text{ ev}, \quad (20)$$

which is consistent with currently evaluated values of γ_1, 0.3 ev–0.5 ev.[16,17]

(2) ω_p^2 for $\Theta=\pi/2$ depends only on γ_0 as

$$\omega_p^2(\pi/2)=\frac{4\pi e^2}{m}n\left[1-\frac{m}{2\hbar^2}a^2\gamma_0\right]. \quad (21)$$

Thus, if $\gamma_0<2\hbar^2/(ma^2)=2.52$ ev, we will observe the energy loss due to the plasma excitation for the over-all range of angle $0<\Theta<\pi/2$, while if γ_0 exceeds the above value, we will get a critical angle Θ_C, above which $\hbar\omega_p$ becomes an imaginary number; thus, there ceases to exist a stable plasma motion of electrons. Figure 1 shows the Θ dependence of $\hbar\omega_p$ for various choices of parameter γ_0. Figure 2 shows how the critical angle Θ_C changes with the γ_0. If we take an average of (18) over the Θ, corresponding to a polycrystal foil, we get

$$\omega_p^2=\frac{4\pi e^2}{m}n\left[1-\frac{ma^2}{3\hbar^2}\gamma_0\left(1+\frac{1}{4}\frac{\gamma_1}{\gamma_0}\left(\frac{c}{a}\right)^2\right)\right]. \quad (22)$$

Since γ_1 is determined to be equal to 0.32 ev from Watanabe's data, Eq. (22) gives γ_0 dependence of the energy loss in polycrystalline foil, which is shown in Fig. 3. Thus, if one measures Θ dependence of $\hbar\omega_p$, using a single-crystal thin foil of graphite, it gives us very useful information about the resonance energy parameters γ_0 and γ_1. So far we have only the experi-

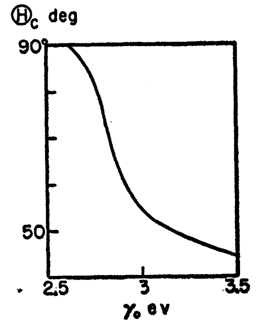

FIG. 2. The γ_0 dependence of the critical angle Θ_C.

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[16] J. Hove, Phys. Rev. **100**, 645 (1956).

[17] K. Komatsu and T. Nagamiya, J. Phys. Soc. Japan **6**, 438 (1951).

ment, which can let us determine γ_1, namely the work done by Watanabe.

Next, we will discuss the θ dependence of the energy loss for the case of $\Theta=0$, which has been measured by Watanabe. As we have shown in the reference 14, the dispersion formula for the eigenfrequency of the collective oscillation is given as follows:

$$\hbar\omega=\hbar\omega_p+\hbar\frac{2\pi}{3}\frac{e^2}{m^2\omega_p{}^3}\frac{1}{V}$$

$$\times\sum_{\mathbf{p}\cdot\lambda}\int d^3xu_n{}^{p*}(x)\nabla^2V(x)u_n{}^p(x)N_n(p,\lambda)$$

$$+\hbar\frac{4\pi e^2}{m^2\omega_p{}^3}\frac{1}{V}\sum_{p\cdot\lambda}\int d^3xu_n{}^{p*}(x)\frac{p^2}{2m}u_n{}^p(x)N(p,\lambda)k^2$$

$$+\hbar\frac{4\pi e^2}{m\omega_p{}^3}n\frac{\hbar^2}{4m^2}k^4, \qquad (23)$$

where $V(x)$ is the potential of atomic nucleus. Hence, by the Poisson's equation, we have

$$\nabla^2V(x)=-4\pi Ze^2\delta(x). \qquad (24)$$

Thus, the second term vanishes for π electrons. Let us consider the k^2 term of Eq. (23),

$$\hbar\frac{4\pi e^2}{m^2\omega_p{}^3}\frac{1}{V}\sum_{p\cdot\lambda}\int d^3xu_n{}^{p*}(x)\frac{p^2}{2m}u_n{}^p(x)N(p,\lambda)k^2$$

$$=2\frac{4\pi e^2}{m\omega_p{}^3}\frac{1}{V}\sum_{p\cdot\lambda}\int d^3xu_n{}^{p*}(x)\frac{p^2}{2m}u_n{}^p(x)N_n(p,\lambda)E\theta^2. \qquad (25)$$

The right-hand side of the above equation is obtained by the substitution of $k=p_0\theta/h$ and E is the kinetic energy of incident electron $p_0{}^2/2m$. To evaluate the average kinetic energy of π electrons, we need to know the wave function of π electrons in graphite. However, since this term is a correction term, we may estimate it in the free-electron approximation with the effective mass given by Eq. (18). Thus, the average kinetic energy of π electrons is given as follows,

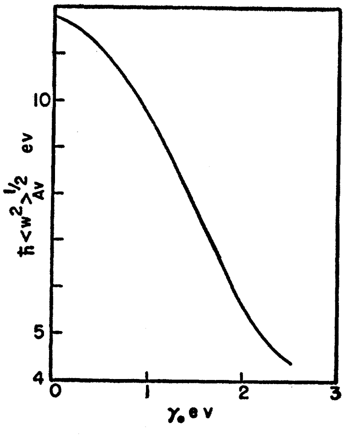

FIG. 3. The γ_0 dependence of the π-electron plasma frequency in polycrystalline graphite.

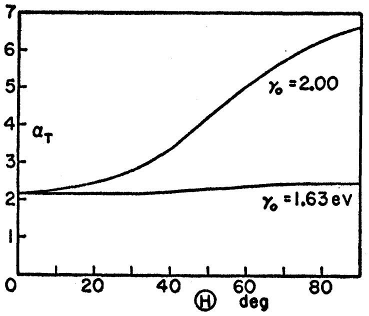

FIG. 4. The Θ dependence of the coefficient α_T.

$$\frac{1}{V}\sum_{\mathbf{p}\cdot\lambda}\int u_n{}^{p*}(x)\frac{p^2}{2m}u_n{}^p(x)d^3xN_n(\mathbf{p},\lambda)=\tfrac{3}{5}\epsilon_0{}'n, \qquad (26)$$

where

$$\epsilon_0{}'=\frac{\hbar^2}{2m(\Theta)^*}\left(\frac{3n}{8\pi}\right)^{\frac{2}{3}}. \qquad (27)$$

Then, we get

$$(25)=\frac{6}{5}\left(\frac{4\pi e^2}{m}n/\omega_p{}^2\right)(\epsilon_0{}'/\hbar\omega_p)E\theta^2. \qquad (28)$$

This gives the following value for the coefficient α_T of $E\theta^2$, for the case of $\Theta=0$,

$$\alpha_T=2.15, \qquad (29)$$

which is in surprisingly good agreement with the experimental value of $\alpha=2.0\pm0.6$. Figure 4 shows the Θ dependence of α_T for the choices of $\gamma_0=1.63$ ev, and 2.0 ev, respectively. Finally, the k^4 term

$$\hbar\frac{4\pi e^2}{m\omega_p{}^3}n\frac{\hbar^2}{4m^2}k^4=\frac{E}{2\hbar\omega_p}\left(\frac{4\pi e^2}{m\omega_p{}^2}\right)nE\theta^4, \qquad (30)$$

gives the following value for the coefficient β_T of $E\theta^4$,

$$\beta_T=5\times10^3 \text{ at } E=25 \text{ kev}, \quad \Theta=0. \qquad (31)$$

This figure might appear to be large enough to be determined from the observed result. However, as one can see from Watanabe's data of graphite, the contribution of the θ^4 term is within the error of the determination of the θ^2 dependence of the energy loss.

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