reduction of the force against the pinning centers and leaves the pulse height unchanged.

VIII. CONCLUSIONS

Transmission of pulsed perpendicular magnetic fields through type-II superconducting plates in a perpendicular magnetic field has been observed. For sufficiently large pulses pinning is insignificant except very close to H_{c1}. This pinning near H_{c1} can be reduced if the applied field is changed so that the change ΔH aids the pulse field. The data can be interpreted in terms of an effective resistivity ρ. The data points are somewhat below a line given by $\rho/\rho_n=(H-H_{c1})/(H_{c2}-H_{c1})$.

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L_2-L_3N Coster-Kronig Transition Probabilities and L-Subshell Fluorescence Yields of Hg and Ta†

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The probability for the radiationless transfer of L_2 vacancies to the L_3 subshells of Hg and Ta has been measured directly, by observing the spectra of L x rays from Tl^{204} and W^{181} in coincidence with K_{α_2} x rays. The L_α component of the coincident spectrum provides a measure of the L_2-L_3N Coster-Kronig transition probability, L_2-L_3M transitions being energetically impossible in this region. The following values are found: for Hg, $f_{L_2L_3}=0.08\pm0.02$; for Ta, $f_{L_2L_3}=0.20\pm0.04$. The x-ray coincidence experiments show that the average number of x-ray quanta emitted after creation of a primary vacancy in the L_2 subshell is $\nu_{L_2}=0.42\pm0.02$ for Hg and $\nu_{L_2}=0.31\pm0.01$ for Ta. Subshell fluorescence yields are found to be $\omega_{L_2}=0.39\pm0.03$ and $\omega_{L_3}=0.40\pm0.02$ for Hg; $\omega_{L_2}=0.25\pm0.02$ and $\omega_{L_3}=0.27\pm0.01$ for Ta.

I. INTRODUCTION

PRECISE L-shell fluorescence yields are needed, particularly in the heavy-element region, to obtain nuclear electron-capture ratios from x-ray intensity measurements. In addition, these yields are of interest in atomic physics. The L-shell fluorescence yield of an element depends upon the relative numbers n_i ($i=1$, 2, 3) of primary vacancies produced in the three L subshells, on the individual subshell fluorescence yields ω_{L_i}, and on the Coster-Kronig transition probabilities $f_{L_iL_j}$ for the radiationless transfer of vacancies between the subshells. The average numbers ν_{L_i} of L x-ray quanta emitted per primary vacancy in the ith subshell are

$$\begin{aligned}\nu_{L_1}&=\omega_{L_1}+f_{L_1L_2}\omega_{L_2}+(f_{L_1L_3}+f_{L_1L_2}f_{L_2L_3})\omega_{L_3},\\ \nu_{L_2}&=\omega_{L_2}+f_{L_2L_3}\omega_{L_3},\qquad (1)\\ \nu_{L_3}&=\omega_{L_3}.\end{aligned}$$

In the shifting of vacancies between L subshells by Coster-Kronig transitions, the difference in binding energies is carried off by an outer shell electron. Consequently, such a transition between two subshells is only possible when the difference in binding energies exceeds the ionization energy of an outer shell. For example, an L_2-L_3M_5 transition is possible for atomic number Z if $E_{L_2}(Z)-E_{L_3}(Z)>E_{M_5}(Z+1)$, assuming that the M binding energy in an atom of atomic number Z with an L vacancy is approximately equal to the M binding energy of a neutral atom of atomic number $Z+1$. The uncertainty inherent in this assumption and in the various subshell binding energies makes it difficult to predict the exact range in atomic numbers where given Coster-Kronig transitions are energetically possible. Approximate ranges in Z for various types of Coster-Kronig transitions have been listed by several authors.[1-3] The probability of energetically possible transitions varies with the phase space available to the ejected electron and with the extent to which its wave function overlaps the initial bound-state wave function.

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This overlap depends on the energy of the ejected electron. There is little overlap when this energy is small or very large. The L_1-$L_{2,3}M_{4,5}$ Coster-Kronig transitions have been the subject of two recent theoretical investigations,[4,5] but there is a lack of theoretical work on transitions of the L_2-L_3 type.

It is apparent from Eqs. (1) that determination of L-subshell fluorescence yields generally requires a knowledge of Coster-Kronig transition probabilities. In most experiments on L fluorescence yields, primary vacancies are found in all three L subshells. Some work has been performed, using an x-ray coincidence method, to obtain one partial L-shell fluorescence yield (ω_{KL}) that results from primary vacancies located in the L_2 and L_3 subshells only.[6–11] Individual subshell fluorescence yields have rarely been measured. The earliest experiments to this effect were performed by Küstner and Arends,[12] and by Stephenson.[13] With suitable incident radiation, these workers produced vacancies in the L_3 subshell alone, or in the L_2 and L_3 subshells, or in all three subshells, and with an ionization chamber compared the intensity of the radiation falling on a radiator with the fluorescence radiation from it. Kinsey[14] has calculated subshell yields by estimating, from the best available measurements, the radiation width of each subshell energy level and dividing it by the total width of the level. Jopson *et al.*[15] have obtained L_3 subshell fluorescence yields through an x-ray coincidence method in which the K_{α_2} component of the K x rays had been eliminated with the help of secondary radiators.

The majority of currently used subshell fluorescence yields and Coster-Kronig transition probabilities have been derived from experimental results on mean L-shell fluorescence yields and L Auger spectra. A detailed example of such a calculation is given by Ross *et al.*[16] In calculations of this type, the simplifying assumption is usually made that the Coster-Kronig transition probability $f_{L_2L_3}$ vanishes in the region from $Z=30$ to $Z=90$. This assumption is based on the fact that, in this region, the binding-energy difference between the L_2 and L_3 subshells is insufficient to permit ejection of an M electron, which would be the most probable process otherwise. It remained to be tested experimentally whether the L_2-L_3N transition rate is indeed negligible and hence, whether the assumption $f_{L_2L_3}\approx 0$ $(30<Z<90)$ is valid. Some results to the contrary have been reported. From an extensive analysis of available data on Bi, Ross *et al.*[16] found $f_{L_2L_3}=0.06\pm0.14$ for $Z=83$. Nall, Baird, and Haynes[17] report $f_{L_2L_3}=0.22\pm0.04$ for Hg, and Sujkowski[18] lists $f_{L_2L_3}=0.25\pm0.13$ for Tl. However, these results have been obtained by assuming certain values for one or the other of the L subshell fluorescence yields. In the present investigation, a direct experimental determination of the L_2-L_3N Coster-Kronig transition probability has been made for Hg and Ta.

II. EXPERIMENTAL METHOD

In principle, the experiment consists in observing the spectrum of L x rays in coincidence with K_{α_2} x rays. The latter indicate the formation of L_2 vacancies. If these vacancies are filled by radiative transitions from outer shells, x rays characteristic of the L_2 subshell are emitted (L_{β_1}, L_{γ_1}, L_{γ_5}, L_{γ_6}, etc.). If, however, some L_2 vacancies are transferred to the L_3 subshell by Coster-Kronig transitions and the latter vacancies are radiatively filled, then the L x-ray spectrum will include components characteristic of the L_3 subshell. In particular, L_α x rays from the most probable L_3-$M_{4,5}$ transitions will be emitted, and the relative intensity of these x rays can be used to deduce the Coster-Kronig transition probability.

For the experiment on Hg, a thin vacuum-evaporated source of Tl^{204} on Mylar backing was employed. The K x-ray detector was a scintillation counter consisting of a 5-mm thick Harshaw NaI(Tl) crystal with a beryllium window, mounted on a Philips XP1010 photomultiplier tube. To select K_{α_2} x rays, tungsten was used as critical absorber, since its K absorption edge falls between the Hg K_{α_1} and K_{α_2} x-ray energies. The fluorescent W K x rays were absorbed in a layer of Er, and additional Sn and Cu absorbers were included to obtain clean Hg K_{α_2} x rays. Figure 1 shows the composite, unfiltered K x-ray spectrum from Tl^{204} and the spectrum of K_{α_2} x rays transmitted by the series of absorbers. Residual Hg K_{α_1} x rays in the filtered K_{α_2} x-ray beam do not exceed one percent of the beam intensity. *Note added in proof.* The purity of the K_{α_2} x rays was verified with a Ge(Li) detector of 3.4-keV resolution. The line shape of these x rays is indistinguishable from the line shape produced by monochromatic gamma rays.

In the work on Ta, a source of carrier-free W^{181} was used, which had been vacuum-evaporated on Mylar. Critical absorption with Er, followed by Sm, Sn, and

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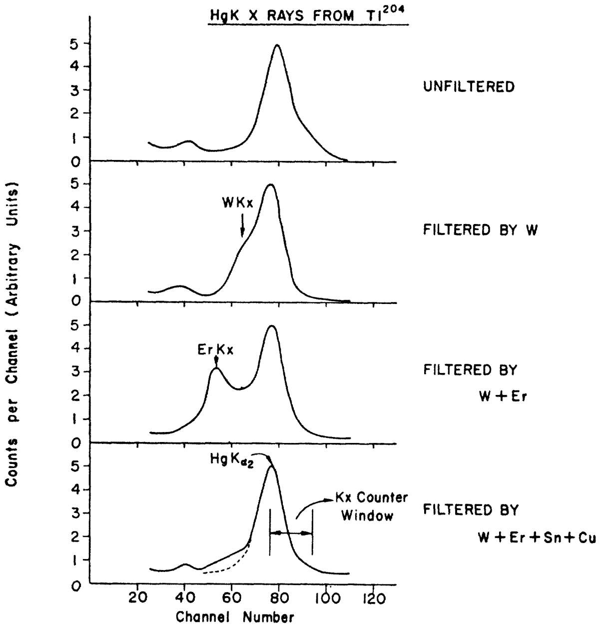

FIG. 1. Spectra of Hg K x rays from Tl^{204}. The K_{α_1} component is removed by critical absorption with W. Fluorescent x rays from W are absorbed in layers of Er, Sn, and Cu.

Cu filters, reduced the intensity of Ta K_{α_1} x rays to less than 3% of the filtered beam intensity.

The L x rays were detected in a proportional counter[11] filled to 1 atm with a 9:1 mixture of argon and methane. The gas was taken through a cold trap before entering the counter. Good stability was attained over the protracted counting periods which were made necessary by low counting rates from the highly collimated L x-ray beam and the severely attenuated K x rays.

Because of the inherent fluctuations in the proportional counter's response time, a slow coincidence resolving time of 2.2 μsec was chosen, lest true coincidence events be missed. Hence, the chance coincidence counting rates were considerable. The total and chance coincidence spectra were therefore recorded simultaneously in the two halves of a 512-channel analyzer memory, employing two identical coincidence units provided with identical input signals, but one of them having a 25-μsec delay introduced in one channel.[19] The coincidence units were interchanged for every other run, in order to cancel out errors from any slight difference between the effective resolving times of the two units.

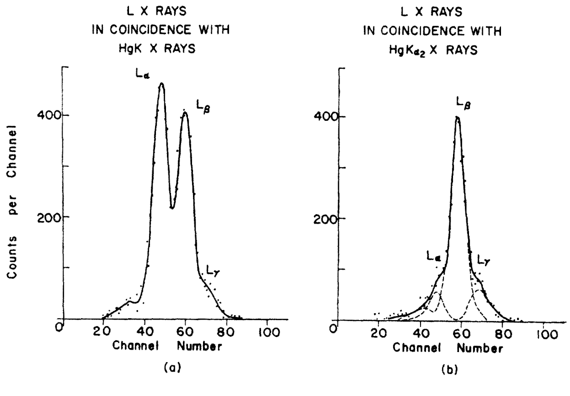

FIG. 2. Typical spectra of Hg L x rays from Tl^{204}, obtained with a proportional counter. Spectrum (a) is gated by unfiltered K x rays, and hence arises from primary vacancies in both the L_2 and L_3 subshells. Spectrum (b), gated by K_{α_2} x rays only, arises from primary vacancies in the L_2 subshell alone; Coster-Kronig transitions account for the residual L_α peak.

Typical Hg L x-ray spectra are shown in Fig. 2, obtained in coincidence with all K x rays and in coincidence with K_{α_2} x rays only. The small L_α peak in the latter spectrum indicates the presence of L_2-L_3 Coster-Kronig transitions. The spectra were analyzed into L_α, L_β, and L_γ components, including their associated argon escape peaks, with the aid of line shapes derived from Ge, Br, and Rb K x rays excited by fluorescence and recorded under the same conditions. Corrections for attenuation and counter efficiency were performed as described elsewhere[11] in order to obtain the intensity ratios $I_{L_\alpha}:I_{L_\beta}:I_{L_\gamma}$, which are listed in Table I.

TABLE I. Relative intensities of L_α, L_β, and L_γ x rays in coincidence with K_α x rays, and average number ν_{L_2} of L x-ray quanta emitted after creation of a primary vacancy in the L_2 subshell.

Z	$I_{L_\alpha}:I_{L_\beta}:I_{L_\gamma}$ for L x rays in coincidence with K_{α_1} and K_{α_2}	K_{α_2} only[a]	ν_{L_2}
73	53.4:40.2:6.4	15.0:74.7:10.3	0.31±0.01
80	52.0:40.0:8.0	6.2:76.9:16.9	0.42±0.02

[a] Corrected for residual K_{α_1} x rays in filtered K x-ray beam.

III. DETERMINATION OF ν_{L_2}

The average number ν_{L_2} of L x-ray quanta emitted in the de-excitation of an atom with an L_2 primary vacancy can be obtained directly from the coincidence counting rate N_c between K_{α_2} and L x rays, because of the relation

$$N_c = N_{K_{\alpha_2}} \epsilon_L a_L \Omega_L \nu_{L_2}. \qquad (2)$$

Here, $N_{K_{\alpha_2}}$ is the net counting rate in the K_{α_2} x-ray detector, ϵ_L and Ω_L are the efficiency and solid angle pertaining to the L x-ray detector, and a_L is the correction factor for absorption of L x rays between source and detector. In computing this absorption correction, the relative intensities of the L_α, L_β, and L_γ components were taken into account.

Since N_c refers to coincidences with *all* L x rays, it was not necessary to resolve the L x-ray spectrum in determining this quantity. The L x-ray detecting proportional counter could, therefore, be replaced with a scintillation counter for this phase of the experiment. A 5-mm thick NaI(Tl) crystal with Be window provided increased efficiency and solid angle and permitted shorter coincidence resolving time.

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Results of the determinations of ν_{L_2} are included in Table I.

IV. DETERMINATION OF $f_{L_2L_3}$, ω_{L_2}, AND ω_{L_3}

The calculations of this section are based on three relations. The first of these expresses the fact that atoms with a primary vacancy in the L_2 subshell will, in the course of de-excitation, emit L_2 or L_3 x rays in the ratio

$$I_{L_3}/I_{L_2}=(f_{L_2L_3}\omega_{L_3})/\omega_{L_2}. \tag{3}$$

The second relation states that the average number ν_{L_2} of L x-ray quanta emitted after creation of a primary vacancy in the L_2 subshell is

$$\nu_{L_2}=\omega_{L_2}+f_{L_2L_3}\omega_{L_3}. \tag{4}$$

The third relation defines the partial fluorescence yield ω_{KL}, which corresponds to the distribution of primary L subshell vacancies following K_α x-ray emission:

$$\omega_{KL}=n_2\nu_{L_2}+n_3\omega_{L_3}. \tag{5}$$

The three equations, (3), (4), (5), can be solved to obtain the Coster-Kronig transition probability $f_{L_2L_3}$ and the subshell fluorescence yields ω_{L_2} and ω_{L_3}, once I_{L_3}/I_{L_2}, ν_{L_2}, and ω_{KL} have been determined.

The ratios I_{L_3}/I_{L_2} have been calculated from the measured intensity ratios $I_{L_\alpha}:I_{L_\beta}:I_{L_\gamma}$ (Sec. II). All L_α x rays and a small fraction of the L_β x rays originate from the filling of L_3 vacancies. The fraction of L_β x rays originating from the L_3 shell has been computed from M. Goldberg's measurements.[20]

An experimental determination of the partial fluorescence yield $\omega_{KL}=0.41\pm0.02$ for Hg has been reported previously.[11] The value of ω_{KL} for Ta has been measured by Jopson *et al.*,[8] who obtained 0.28±0.01. A redetermination of this fluorescence yield was performed as part of the present work, with the identical result. The ratio $n_2:n_3$ of primary vacancies following K x-ray emission is equal to the intensity ratio of K_{α_2} to K_{α_1} x rays, tabulated by Nijgh *et al.*[21]

Table II contains a summary of the final results.

TABLE II. Measured L_2 and L_3 subshell fluorescence yields and L_2-L_3 Coster-Kronig transition probabilities.

Element	ω_{L_2}	ω_{L_3}	$f_{L_2L_3}$
$_{73}$Ta	0.25±0.02	0.27±0.01	0.20±0.04
$_{80}$Hg	0.39±0.03	0.40±0.02	0.08±0.02

V. DISCUSSION

The L subshell fluorescence yields of Hg and Ta obtained in the present work differ from values calculated under the assumption that the L_2-L_3 Coster-Kronig transition probability can be neglected.[15] The subshell yields ω_{L_2} reported elsewhere under the assumption $f_{L_2L_3}\approx0$ are actually the average number ν_{L_2} of L x-ray quanta emitted after creation of a primary vacancy in the L_2 subshell; this number is greater than ω_{L_3}. However, $\nu_{L_2}\neq\omega_{L_2}$ when $f_{L_2L_3}\neq0$ [cf. Eqs. (1)]. When the nonvanishing L_2-L_3 Coster-Kronig probability is taken into account, it is found that ω_{L_2} is smaller than ω_{L_3} in the region under consideration. This result is contrary to the general trend assumed, for example, in the review article by Listengarten,[3] but it is not unreasonable: x-ray emission and Coster-Kronig transitions (in addition to the ordinary Auger process) are alternative modes of deexcitation for an atom ionized in the L_2 subshell, while only x-ray and ordinary Auger-electron emission compete when an L_3 vacancy is to be filled.

The L_2-L_3N Coster-Kronig transition probability is found to be smaller for Hg than for Ta. This fact is in qualitative agreement with the trend to be expected, since the N electron ejected from Hg has higher energy (1.1 to 1.5 keV, depending on the subshell) than the N electron ejected from Ta (0.7 to 1.0 keV), and the wave length of these electrons is, in either case, substantially shorter than the radial distance over which the bound-state N-electron wave functions have an appreciable magnitude.[22]

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