# Lifetimes of the  $2^{1}$ Ne 2796-, 1747-, and 2867-keV levels

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The mean lives of the <sup>21</sup>Ne  $1/2$ <sup>+</sup>,  $7/2$ <sup>+</sup>, and  $9/2$ <sup>+</sup> levels at 2796, 1747, and 2867 keV have been measured using the  ${}^{4}$ He( ${}^{18}$ O,*n*)<sup>21</sup>Ne reaction near threshold and the Doppler shift attenuation method. The results were  $7.9 + 1.0$ ,  $74 + 6$ , and  $61 + 6$  fs, respectively. In order to obtain a result for the 8-fs 2796-keV level, four different targets were used. These consisted of 'He implanted in thick backings of Mg, Al, Ta, and Au. The analysis developed for measuring this fast lifetime is described in detail. Excitation energies of  $1747.2+0.3$ ,  $2788.87 + 0.10$ ,  $2795.8 + 0.7$ , and  $2867.2 + 0.4$  keV were obtained for the second-to-fifth excited states. The lifetime results are compared to recent shell-model predictions.

NUCLEAR REACTIONS  ${}^{4}$ He( ${}^{18}$ O,n) ${}^{21}$ Ne,  $E$ =20-27 MeV; measured Doppler shift attenuation and lineshape.  $^{21}$ Ne 2796-, 1747-, and 2867-keV levels; deduced  $\tau_m$ . Implanted targets.

# I. INTRODUCTION

It has recently<sup>1,2</sup> been pointed out that the  $J^{\pi}$  $=\frac{1}{2}$ ,  $\frac{1}{2}$  doublet in <sup>21</sup>Ne at 2789 and 2796 keV provides a very favorable possibility for the study of the parity nonconserving weak interaction in the nucleus. .

Such parity mixing would result in a nonzero circular polarization  $(P_{\nu})$  for the 2789-keV groundstate decay (see Fig. 1), and a recent measurement<sup>3</sup> put an upper limit of 0.5% on  $P_{\gamma}$ . An alternative method to investigate such mixing would be to measure the asymmetry  $(A<sub>x</sub>)$  of the 2789-keV ground-state decay following formation of this level using a polarized beam. To obtain the parity mixing matrix element from either  $P_r$  or  $A_r$ , a constant of proportionality<sup>4</sup> must be evaluated which requires knowledge of  $\tau(M1)$ , the partial mean life of the M1 part of the  $\frac{1}{2}$  +  $\frac{3}{2}$  + 2796 – 0 transition. The 2796-keV level. decays predominantly to the ground state, and as the  $E2$  contrimantly to the ground state, and as the  $E_2$  contri-<br>bution is negligible,<sup>4</sup> the measurement of the total mean life of this level allows an evaluation of this constant which is necessary for present and future parity nonconserving experiments in  $2^1$ Ne.

An accurate measurement of this lifetime is discussed in this paper. The 2796-keV level was known to have a mean life of  $\tau < 20$  fs<sup>5-7</sup> so that an evaluation of the required accuracy involved using the Doppler shift attenuation (DSA) method at the limit of its applicability and also the development of new facets of this technique. Thus a detailed description of the experimental method and analysis of data has been given with emphasis on the many factors which must be taken into consideration in such a measurement. The precision obtained is competitive with the most accurate

previous results in the 1-10 fs lifetime range.

The lifetimes of the 1747- and 2867-keV levels (see Fig. 1) also reported herein are considerably (see Fig. 1) also reported herein are consideral longer $5,6,8-11$  than that of the 2796-keV level and could thus be extracted using well established  $\gamma$ could thus be extracted using well established<br>ray line shape fitting techniques.<sup>12-14</sup> However they are also sufficiently short-lived to be analyzed by the technique developed for the 2796-keV level, and permit a convenient comparison between the two methods.

The stopping power of ions in matter provides the most important ancillary input to any DSA measurement. In the next section we present the data and formalism used for the stopping of  $^{21}$ Ne ions in matter and then describe the DSA methods applicable in the present analysis. Some further details of the stopping power are discussed in an Appendix. In Sec. III the experimental results and their analysis are described. Finally, the methods used and the results obtained are compared to previous work in Sec. IV and matrix elements extracted from the experimental lifetimes are compared to shell-model predictions.

### II. THE DSA ANALYSIS

# A. The specific energy loss of  $2<sup>1</sup>$  Ne and its parametrization

The DSA measurements for the <sup>21</sup>Ne 2796-keV level were performed at a  $2N$ Ne recoil velocity of  $\beta$ ( $\equiv v/c$ )  $\approx$  0.04. Thus, accurate results for the energy loss are desired near this velocity and, to a lesser extent, at lower velocities. We consider the specific energy loss  $dE/d(\rho x)$  of an ion with nuclear charge  $Z_1$ , mass  $A_1$ , and velocity v, in a material with nuclear charge  $Z_2$ , mass  $A_2$ , and density  $\rho$ .  $dE/d(\rho x)$  is composed of a nuclear part, only important at low velocities  $(v/v_0 \le 1,$ 

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FIG. 1. The <sup>21</sup>Ne energy levels for  $E_x < 3.6$  MeV and the  $\gamma$ -ray transitions pertinent to the present study. The level energies (in keV) are from the present study except for the first (Ref. 5) and the fifth—which is based on the energy of the  $2.87 \rightarrow 1.75$  transition (Ref. 5) and the present excitation energy of the 1.75-MeV level. The spin-parity assignments and branching ratios are from Ref. 5. The numbers in parentheses are the uncertainties in the least significant figure.

where  $v_0 = c/137$ , and an electronic part. The specific energy loss of  ${}^{21}$ Ne ions was obtained from a mixture of empirical and semi-empirical results. Porat and Ramavataram<sup>15</sup> provided accurate  $(+5\%)$  electronic energy loss data for Ne ions stopping in Al, Ni, and Au for the velocity range  $0.9 \le v/v_0 \le 3.52$ . For Ne projectiles, this data provided the main input for Northcliffe and Schilling's encyclopedic tabulation<sup>16</sup> of semi-empirical stopping powers. For Al, further data in the range  $6.4 < v/v_0 < 20$  was provided by North-<br>cliffe.<sup>17</sup> For Ne in Al the tabulation of Northcl cliffe.<sup>17</sup> For Ne in Al the tabulation of Northcliff and Schilling is quite reliable since, in their semiempirical fits, stopping powers for various ions

in Al provided the master reference curves for interpolation to other projectiles and materials. Because the experimental results of Porat and Bamavataram provided direct input, the stoppingpower curves of Northcliffe and Schilling for Ne ions in Ni and Au are also relatively reliable. In recent years increasingly detailed and accurate stopping power data have revealed departures from the smooth dependence of  $dE/d(\rho x)$  on  $Z_1$  and  $Z_2$ assumed by Northcliffe and Schilling. There exist several quite successful effective charge prescriptions for taking these oscillations into account in the interpolation procedures necessary to obtain stopping-power curves where no experimental data exist. We have considered four of these in order to obtain the best results for Ne ions in Mg and Ta—for which no direct measurements exist—and to check the interpolation of Northcliffe and Schilling for Ne in Ni and Au. All four procedures rely on the assumption that for two ions  $a$  and  $b$  at the same velocity in the same stopping material, the stopping powers are related by

$$
\frac{1}{(\gamma Z_1)_b^2} \left( \frac{dE}{d(\rho x)} \right)_b = \frac{1}{(\gamma Z_1)_a^2} \left( \frac{dE}{d(\rho x)} \right)_a, \tag{1}
$$

where  $\gamma$ , the effective charge parameter, is assumed to depend on  $v$  and  $Z$ , only. The four procedures used are as follows:

(i) Ward. Ward et  $al.^{18}$  constructed a careful parametrization of effective charge good for  $v/v_0$  $\geq$  2. Following their prescription, the desired stopping-power curve for a given ion may be obtained from an accurate table of 'He stopping-power data for that material, using their effective charge parametrization and the relationship given by Eq. (1). We have followed this procedure to construct a computer program based on the  ${}^{4}$ He tables of Ziegler and Chu.<sup>19</sup> tables of Ziegler and Chu. bles of Ziegler and Chu.<sup>19</sup><br>(*ii*) Forster. Forster et al.<sup>20</sup> made measure

ments for  $Z_1 = 9$ , 12, 13, 16, and 17 stopping in elements with  $Z_2 = 22$ , 26, 28, 29, 47, and 79. They then used the effective charge parametrization of (i) to interpolate to other values of  $Z<sub>1</sub>$  for the above values of  $Z_2$ .

e above values of  $Z_2$ .<br>(*iii*) Schwalm. Schwalm et al.<sup>21</sup> developed an approach utilizing measurements of  $dE/d(\rho x)$  for protons and an associated analytical representation of the effective charge. The Schwalm parametrization is especially adapted to  $10 \le Z_1 \le 17$ and  $11 \leq Z_2 \leq 17$ .

(iv) Braune. Braune<sup>22</sup> combined the effective charge parametrization of (i) with an analytic representation of a "universal materials" function  $[F(Z_2;Z(V))$  in Eq. (22) of Ref. 21] which quite successfully reproduces the  $Z<sub>2</sub>$  oscillations described, e.g., in Ref. 18.

The predictions of  $(i)$ - $(iv)$ , and of Northcliffe



	Forster <sup>a</sup>	Ward <sup>b</sup>	Energy loss (MeV $\text{cm}^2/\text{mg}$ ) Schwalm <sup>c</sup>			
Stopping material				Braune <sup>d</sup>	Northcliffe <sup>e</sup>	Average <sup>r</sup>
Mg	$\cdots$	10.67	9.74	10.11	$\cdots$	10.17(4.6)
Al	$\cdots$	9.26	8.93	9.43	9.58	9,30(3,0)
Ni	6.56	6.96	6.67	6.95	6.62	6.75(2.8)
Ta	$\cdots$	3.50	3.49	3.48	3.50	3.49(0.3)
Au	3.62	3.62	3.34	3.31	3.31	3.44(4.8)

TABLE I. Comparison of various effective charge prescriptions for  $-dE/d(\rho x)$  for <sup>21</sup>Ne ions at  $v/v_0 = 5.5$ .

 $a$ Reference 20.

<sup>b</sup> Reference 18.

 $c$ Reference 21.

 $d$ Reference 22.

Reference 16.

<sup>f</sup> The numbers in parentheses give the internal errors in percent.

and Schilling for Ne ions of velocity  $v/v_0 = 5.5$  ( $\beta$ )  $\simeq$  0.04) in five materials are compared in Table I. The results are in remarkably good agreementthe averages all have internal errors of  $<5\%$ . For lower velocities, however, the five  $dE/d(\rho x)$ curves constructed for each stopping material are not in such good agreement. Upon consideration of the various curves it was dec ided that a representation as good as any is that of Northcliffe and Schilling which, after all, is most directly related Schilling which, after all, is most directly relate<br>to the data of Porat and Ramavataram.<sup>15</sup> Accord ingly, the values of  $dE/d(\rho x)$  used for stopping in Al, Ni, Ta, and Au were the curves of Northcliffe and Schilling scaled to pass through the average values of Table I at  $v/v_0 = 5.5$ . For stopping in Mg, the ratio of  $dE/d(\rho x)$  for <sup>20</sup>Ne in Mg and Al,<br>as obtained by the predictions of Schwalm *et al.*<sup>21</sup> as obtained by the predictions of Schwalm et  $al.$ <sup>21</sup>, was used to interpolate the results of Northcliffe and Schilling from  $Z_2 = 13$  to  $Z_2 = 12$ .

The above approach gives the electronic energy loss only. Following procedures developed previously at this laboratory<sup>13,23-25</sup> we parametrize the total specific energy loss as

$$
-M\frac{dv_z}{dt} = \frac{-dE}{d(\rho x)} = K_n(v/v_0)^{-1} + K_e(v/v_0)
$$

$$
-K_3(v/v_0)^3
$$
(2a)

for  $0 \le v \le v_c$  and

$$
-M\frac{dv_z}{dt} \equiv \frac{-dE}{d(\rho x)} = A + B(v/v_0) - C(v/v_0)^2
$$
 (2b)

for  $v > v_c$ , where the initial direction of the recoiling ion is taken as the  $z$  axis. Following previous ing ion is taken as the *z* axis. Following previous considerations for similar kinematical conditions,<sup>21</sup> we chose  $K_n$  equal to 1.26 times the Bohr estimate<sup>26</sup> of the nuclear energy loss evaluated at  $v/v_0 = 1$ . It should be kept in mind that  $K_n(v/v_0)^{-1}$ , which represents the nuclear stopping power, is intended to simulate the change with time of both the magni-

tude and direction of  $v$ . Thus, throughout the remainder of this paper it is understood that  $\nu$  and  $v_z$  are interchangeable. The parameters  $v_c/v_o$ ,  $K_e$ ,  $K_3$ , A, B, and C were varied to obtain the best fit to the data just described with the constraint that both  $-dE/d(\rho x)$  and its derivative were continuous at  $v = v_c$ . Data for  $v/v_0 < 1.9$  were excluded from the fit because this region is not important for present purposes and because the presence of  $Z<sub>1</sub>$  oscillations at low velocities render the interpolation procedures relatively unreliable in this velocity region. The results of the least squares fits are displayed in Table II. With the exception of  $v_c/v_0$  (dimensionless), the constants of Eq. (2) appearing in Table II are all in units of MeV  $\text{cm}^2/$ mg.

A quantity more directly applicable to the DSA than  $dE/d(\rho x)$  is  $d(v/v_0)/dt$  which we term the slowing power. We obtain [see Eq.  $(2)$ ]

$$
\frac{d(v/v_0)}{dt} (\text{ps}^{-1}) = 4.4105(\rho/A_1) \frac{dE}{d(\rho x)} \left[ \frac{\text{MeV cm}^2}{\text{mg}} \right], \quad (3)
$$

with  $\rho$  in  $g/cm^3$  and  $A_1$  in u. In Fig. 2 curves of  $d(v/v_0)/dt$  for <sup>21</sup>Ne in Mg, Al, Ta, and Au are shown for the  $dE/d(\rho x)$  of Table II. Also shown in Fig. 2 is the relationship between flight time and  $v/v_0$  for an initial velocity  $v_y/v_0 = 5.5$ . The four stopping materials of Fig. 2 constitute a convenient set which encompasses a wide range of slowing powers. For example, at  $v/v_0 = 5.5$  the slowing powers are in the ratio Mg:Al: Ta: Au  $= 1:1.42:3.34:3.82 \text{ ps}^{-1}.$ 

### B. Application of the DSA to fast lifetimes

We wish to develop a simple formalism useful in understanding the application of the DSA to very fast lifetimes and applicable to the present measurement of the 2796-keV level. The concepts are quite similar to a previous treatment of Branford

Stopping material	$K_n^{\mathbf{a}}$	$K_e$	$K_3$	$v_c/v_0$	A	$\boldsymbol{B}$	C	$\epsilon$ (%) b	Range of $v/v_0$ <sup>c</sup>
Mg	0.378	3.310	0.050	4.940	11,006	0.126	0.051	0.8	$1.9 - 9.0$
Al	0.354	3,113	0.051	4.534	8.118	0.639	0.078	0.7	$1.9 - 9.0$
Ni	0.287	1,766	0.000	2.300	$-1.181$	2.956	0.271	0.9	$1.9 - 6.3$
Ta	0.141	0.801	0.000	3.000	$-1.131$	1.601	0.136	1.6	$2.0 - 6.3$
Au	0.134	0.781	0.000	3.000	$-1.072$	1.541	0.129	1.9	$2.0 - 6.3$

TABLE II. Parameters for the energy loss of Ne ions in five materials. The parametrization is given in the text.

<sup>a</sup> This is the only parameter which depends on the atomic weight of the stopping ion. The numbers are for  $^{21}$ Ne. <sup>b</sup> If a constant uncertainty of  $\epsilon$  is assigned to the tabulation of Ref. 16, then a normalized  $\chi^2$  of unity is obtained in the

fit to Eq.  $(2)$ .

<sup>c</sup> The region of  $v/v_0$  included in the fit for <sup>21</sup>Ne.

and Wright. $27$  We consider an idealized monoenergetic, unidirectional beam of excited nuclei traveling in the  $z$  direction with initial velocity  $v(0)$ . The spreads in energy and direction which are inevitably encountered in practice are treated by assuming a skewed Gaussian distribution in velocity with the full width at half maximum (FWHM) and the skewness evaluated from the experimental conditions. We also assume a simple stopping medium, i.e., either stopping in the target or after exiting from a negligibly thin target. Deviations from this assumption are discussed in the Appendix. As the nuclei slow down they decay at the rate

$$
dN(t)/dt = -(N_0/\tau)\exp(-t/\tau) , \qquad (4)
$$



FIG. 2. Slowing power curves for  $^{21}$ Ne ions in four materials. The slowing power is defined by Eq. (3). The top scale for *V* assumes an initial velocity  $v_i/v_0=5.5$ . For each curve the velocities corresponding to transit times of 0, 50, 100, 150, and 200 fs are indicated by vertical scribe marks. The effect of the magnitude of the slowing power on the rate of slowing down is appar-. ent.

and there will thus be a continuous distribution of  $\gamma$ -ray energies  $E_{\gamma}(t)$  between  $E_{\gamma 0}[\equiv E_{\gamma}(\infty)]$  and  $E_{\gamma}(0)$ , where  $E_{\gamma 0}$  is the energy of  $\gamma$  rays emitted by nuclei at rest, and  $E_r(0)$  is the energy of  $\gamma$  rays emitted at  $t=0$  at an angle  $\theta_{\gamma}$  to the z axis. The  $E_{n}(t)$  are given by the Doppler relation:

$$
E_{\gamma}(t) = E_{\gamma 0} \left[ \left[ 1 - \beta(t)^2 \right]^{1/2} / \left[ 1 - \beta(t) \langle \cos \theta_{\gamma} \rangle \right] \right], \qquad (5)
$$

where  $\beta(t) = v(t)/c$  and  $\langle \cos \theta_{\gamma} \rangle$  is the mean value of  $\cos\theta_{\rm w}$ . The extraction of the mean life  $\tau$  from this  $\cos\theta_{\gamma}$ . The extraction of the mean life  $\tau$  from this line shape has been fully discussed previously.<sup>12-14</sup> The procedure used at this laboratory is based on the parametrization for  $dE/d(\rho x)$  of Eq. (2) and is exemplified in Sec. III. Note that to first order Eq. (5) gives

$$
E_{\gamma}(t) \simeq E_{\gamma 0} \left[ 1 + \beta(t) \langle \cos \theta_{\gamma} \rangle \right],\tag{6}
$$

and so, to a very good approximation, the velocity line shape is also the  $\gamma$ -ray energy line shape and is directly observed.

For lifetimes very short compared to the stopping time of the ions, we can take the energy loss as constant,

$$
-dE/d(\rho x) = -Md\nu/dt = K_c(\nu_i/\nu_0), \qquad (7)
$$

where the factorization of the constant to display the initial velocity  $[v_i \equiv v(0)]$  is a convenience as should become apparent. Solving this differential equation gives

$$
V = 1 - \alpha_c^{-1} t \tag{8}
$$

where  $V \equiv v(t)/v(0)$  ranges from 1 to 0 as t varies from 0 to  $\infty$  and  $\alpha_c = Mv_0/\rho K_c$  is the characteristic slowing down time for the energy loss of Eq. (7). The velocity line shape corresponding to the energy. loss of Eq. (7) is obtained using Eq. (4):

$$
dN(V)/dV = x[e^{-x(1-V)} + x^{-1}\delta(V)e^{-x}], \qquad (9)
$$

where  $x = \alpha_c / \tau$ . The ratio of the average Doppler shift to the maximum shift is termed the attenuation factor and is given by

$$
F(\tau) = \int_0^1 V[dN(V)/dV]dV \int_0^1 [dN(V)/dV]dV
$$
  
= 1 - x<sup>-1</sup>(1 - e<sup>-x</sup>). (10)

For  $\tau \ll \alpha_c$ , i.e., large x, we have the useful relation

ation  
\n
$$
1 - F(\tau) = x^{-1} = \tau/\alpha_c,
$$
\n(11)

which is, of course, true for any realistic expression for  $-dE/d(\rho x)$ . Note that the constant energy loss of Eq.  $(7)$  corresponds to the slowing power  $[cf., Eq. (3)]$ 

$$
d(v/v_0)/dt = \alpha_c^{-1}(v_i/v_0), \qquad (12)
$$

with  $\alpha_c = 227A_1/(\rho K_c)$  fs.

It is instructive to consider the very close analogy between the line shape of Eq. (9) and the distribution of time delays  $\psi(t)$  encountered in an electronic timing measurement for a state of mean life  $\tau$ . Such a timing measurement will have a prompt resolution function  $P(t)$ , and  $\psi(t)$  is given by the convolution integral of  $P(t)$  with the exponent:ial decay [for a more detailed discussion of  $\psi(t)$ —usually termed  $F(t)$ —and  $P(t)$  see Ref. 12]

$$
\psi(t) = \int_0^\infty \tau^{-1} \left[ \exp(-t'/\tau) \right] P(t-t') dt' . \tag{13}
$$

Similarly, the experimentally observed line shape corresponding to the theoretical line shape of Eq. (9) will be

$$
[dN(V)/dV]_{\text{exp}} = \alpha_c \int_0^1 \tau^{-1} {\exp[-\alpha_c(1 - V')/\tau] } \times \phi(V - V')dV', \qquad (14)
$$

where  $\phi(V - V')$  is the detector response function. We have suppressed the  $\delta$  function part of Eq. (9) because it is negligible for  $\tau \ll \alpha_c$  and, in any case, not relevant to the present discussion. It is seen that the analogy between the time distribution of Eq.  $(13)$  and the line shape of Eq.  $(14)$  is exact if we make the connection of variables between Eqs. (13) and (14) of  $t-1-V$ ,  $\tau-\tau/\alpha_c$ . [A more natural but completely equivalent connection would be  $t \rightarrow \alpha_c(1 - V)$ ,  $\tau \rightarrow \tau$ . Once this connection is made it is a simple matter to apply the expertise and mathematical techniques developed for electronic timing to the DSA.. For instance, there are two methods for extracting the lifetime from the distribution of time delays,  $\psi(t)$ , namely the slope and the centroid methods. In general, the slope method is more accurate but becomes less so as  $\tau$  decreases until below some boundary value of  $\tau$ , say  $\tau_{\textit{b}}^{\vphantom{\dagger}}$  it becomes more accurate to determine  $\tau$ from the centroid method. Likewise, lifetimes are extracted from the DSA by either line shape fitting or from the centroid of the line shape, i.e.,



FIG. 3. Line shapes for  $2^{1}$ Ne decaying in Au. The theoretical line shape corresponding to the  $-dE/d(\rho x)$  of Eq. (2) and Table II has been convoluted with the assumed detector response function indicated by the dashed curve. The resulting line shapes (solid curves) for  $\tau = 5$ , 20, and 80 fs are indicated. The intensity scale for each is arbitrary. For 20 and 80 fs the dot-dashed curves have logarithmic slopes corresponding to 20 and 80 fs, respectively, and thus correspond to Eq. (14), i.e., to the assumption of a constant  $-dE/d(\rho x)$ . For 5 fs the dotdashed and solid curves are undistinguishable. The logarithmic slopes were calculated using  $\alpha_c$  = 384 fs.

from  $F(\tau)$  vis  $\tilde{a}$  vis Eq. (11), and we wish to utilize results familiar from electronic timing to determine  $\tau_h$  for the DSA.

Figure 3 shows line shapes for  ${}^{21}$ Ne decaying in Au with  $\tau=5$ , 20, and 80 fs. The solid curves are the line shapes for the  $dE/d(\rho x)$  of Eq. (2) as dethe line shapes for the  $dE/d(\rho x)$  of Eq. (2) as described by Warburton *et al.*<sup>13</sup> They were obtained by convolution with the Gaussian (assumed) detector response function illustrated by the dotted curve. First consider the fast lifetime limit for  $dN(V)/dV$  given in Eq. (14). By analogy to electronic timing, we have $^{28}$ 

$$
\frac{d}{d(1-V)}\left[\ln G(V)\right] = -\frac{\alpha_c}{\tau} \left[1 - \phi(V)/G(V)\right],
$$

where  $G(V)$  is shorthand for  $\left[dN(V)/dV\right]_{\text{exp}}$  and  $\phi(V)$  in this case is given by  $\phi(V)$  $=\exp[-0.5(1 - V)^2/\sigma^2]$ . Thus in the velocity region where  $\phi(V) \ll G(V)$ , the logarithmic slope is a straight line and yields the mean life. Even if the assumption that  $dE/d(\rho x)$  is constant breaks down (as is clearly the case for  $\tau \ge 20$  fs in Fig. 3), we still can say that  $[dN(V)/dV]_{\text{exp}}$  will yield accurate mean lives in the region of velocity where  $\phi(V)$  $\ll G(V)$ . Once again, in analogy with electronic timing, we expect to be able to extract  $\tau$  from the line shape if  $\alpha_c/\tau$  is  $\geq 1.3$  times the slope of the line shape if  $\alpha_c/\tau$  is  $\ge 1.3$  times the slope of the detector response function.<sup>12</sup> For a Gaussian response function, this criterion yields the boundary

value of  $\tau$ 

$$
\tau_b \simeq 0.5 \, (\text{FWHM}/\Delta E_\nu) \alpha_c \,, \tag{15}
$$

below which the mean life is most accurately obtainable from the centroid shift. In Eg. (15), the 'slope of the Gaussian is evaluated at  $\frac{1}{2}$  maximun and the FWHM is in keV as is the full energy shift  $\Delta E_z = E_{\nu}(0) - E_{\nu}(\infty)$ . For Fig. 3, where FWHM = 6 keV and  $\Delta E_r = 112$  keV, we have  $\tau_r \approx 10$  fs.

# III. RESULTS

### A. The general experimental procedure

The inverse reaction  ${}^{4}He({}^{18}O, n\gamma){}^{21}Ne$  ( $Q_0 = -696$ ) keV) was used to form the  $^{21}$ Ne levels under study. Targets were prepared by implanting 'He' ions of 30-40 keV in thick blanks of Mg, Al, Ta, and Au to an areal density of  $\sim$ 4  $\mu$ g/cm<sup>2</sup> as described in the Appendix. The use of 'He-implanted targets in lifetime measurements via DSA line shape analysis has been developed and described by Forster and his co-workers at the Chalk River Laboratories<sup>14</sup> and applications to the measurement of the first-excited state of  $^{22}$ Ne have been made at sevfirst-excited state of <sup>22</sup>Ne have been made at se<br>eral laboratories.<sup>29-31</sup> However, only recently<sup>3</sup> has the method been applied to mean lives less than 100 fs. In a <sup>4</sup>He( $^{19}F, p\gamma$ )<sup>22</sup>Ne study of levels in  $^{22}$ Ne up to 6817 keV,  $\gamma$  rays were observed<sup>32</sup> in coincidence with outgoing particles (as in all previous applications<sup>14,29-31</sup>) and only one stopping medium, Cu, was used. In rough conformity with the estimate given in Sec. II  $[Eq. (15)]$ , a value of 15 fs was found for the fastest lifetime which could be measured in this DSA line shape study.

The present measurements differ from the <sup>4</sup>He(<sup>19</sup>F,  $p\gamma$ )<sup>22</sup>Ne measurements<sup>32</sup> just described in two important details. First, the recoiling  $2N$ e ions were confined to a narrow cone near 0' and feeding from higher-lying levels was avoided by initiating the reaction near threshold rather than by observing  $\gamma$  rays in coincidence with light reaction particles. Second, by using four targets encompassing a wide range of stopping times, a relatively accurate lifetime could be obtained for the fast 2796-keV level  $[\tau < \tau_b$  see Eq. (15)] from centroid rather than line shape analysis.

#### B. The 2796-keV level

#### 1. The kinematics

Thin target excitation functions were recently obtained for formation of the 2789- and 2796-keV<br>levels of <sup>21</sup>Ne via the <sup>18</sup>O( $\alpha$ , *nr*)<sup>21</sup>Ne reaction.<sup>33</sup> levels of <sup>21</sup>Ne via the <sup>18</sup>O( $\alpha$ , n $\gamma$ )<sup>21</sup>Ne reaction.<sup>33</sup> For the 2796-keV level the yield rises from the threshold at  $E_{\alpha}$  = 4.27 MeV to a strong complex resonance with a mean energy,  $E_{\alpha} = 4.406$  MeV and an effective width of  $~125$  keV, and then falls to

essentially a negligible cross section for  $E_{\omega}$  $= 4.46 - 4.56$  MeV before rising again. The complex resonance is well fitted by a sum of two Breit-Wigner resonances with  $(E_{R_1}, \Gamma_1, \sigma_{R_1}) = (4.384 \text{ MeV},$ 82.6 keV, 6.04 mb);  $(E_{R2}, \Gamma_2, \sigma_{R2}) = (4.441 \text{ MeV},$ 30.2 keV, 10.1 mb). For bombardment of  ${}^{4}$ He by  ${}^{18}$ O the resonance energy parameters for the  ${}^{18}$ O rest frame are multiplied by  $\frac{18}{4}$  = 4.5 and become  $(E_{R1}, \Gamma_1) = (19.730 \text{ MeV}, 372 \text{ keV}); (E_{R2}, \Gamma_2)$  $=(19.984 \text{ MeV}, 136 \text{ keV}),$  with a mean energy and effective width of  $(E_R, \Gamma) = (19.825 \text{ MeV}, 560 \text{ keV})$ respectively.

In order to interpret the DSA measurements it is necessary to know the mean energy at which the reaction takes place in the 'He-implanted targets. (Some knowledge of the distribution of reaction energies is also desirable; however, this distribution contributes a minor amount to the total velocity distribution of the recoiling  $2N$ e ions and so is not important.) The mean reaction energy is given by an energy-weighted convolution integral of the resonance with the 'He density distribution. The depth of the 'He in the targets and its density distribution were determined by excitation func-'tions for the  $\frac{1}{2}$  2789-keV level. Details of these measurements and of the determination of the mean reaction energy are given in the Appendix, we only outline the procedures here.

Two sets of targets and of DSA measurements were made. In the first the targets were all made by implantation of 40-keV 'He' ions and the same <sup>18</sup>O bombarding energy,  $E(^{18}O) = 20.1$  MeV, was used for all. For this set the mean reaction energy varied over 180 keV. In principle, a simple correction can be made for this variation; however, some systematic errors may be introduced by this procedure and so a second set of targets was made with the implantation energy adjusted in an attempt to obtain the same mean reaction energy for all targets. The subsequent excitation functions showed that this was not quite obtained; however, it was nearly accomplished by a small variation in bombarding energy from target to target. Finally, the very small adjustments needed to attain data corresponding to a mean reaction energy of 19.825 MeV were made via  $dE_{\gamma}/(E_{\gamma}-E_{\gamma0})$  $\approx dv/v \approx 0.5dE_p/E_p$ . This relationship was also used to adjust the results from the first set of targets.

The  $\gamma$  rays from the <sup>21</sup>Ne 2796  $\div$  0 transition were detected by a 48.5-mm-diameter Ge(Li) detector at  $0^{\circ}$  to the beam axis and with its front face 10.5 cm from the target. The effective mean velocity of the recoiling  $a^2N e^*$  ions is given by

$$
\beta(0) = \beta_{c_{\bullet} m_{\bullet}} (1 + \gamma^{-1} \langle \cos \theta_{c_{\bullet} m_{\bullet}} \rangle) \langle \cos \theta_{\gamma} \rangle, \qquad (16)
$$

where  $\beta_{c.m.}$  and  $cos \theta_{c.m.}$  are the velocity of the center of mass and the angle of the recoiling  $2N$ e ions

in the center of mass, respectively. With the kinematical and experimental conditions described above, we obtain 800—

 $\beta_{c.m.} = (3.978 \pm 0.028)\%$ ,  $\gamma^{-1} = 0.0183 \pm 0.0043$ . (17)

where the indicated excursions in  $\beta_{\rm c.m.}$  and  $\gamma^{-1}$  are those corresponding to a resonance width of 560 keV and are not uncertainties.

Averaging the  $\gamma$ -ray detection angle over the acceptance angle of the  $\gamma$ -ray detector gives  $\langle \cos \theta_{\alpha} \rangle$  $= 0.987 \pm 0.005.^{34}$  Inserting this value of  $\langle \cos \theta_{\gamma} \rangle$  in Eq. (16) and assuming the reaction is symmetric about 90° in the center of mass with  $\beta_{c.m.}$  from Eq. (17), yields

$$
\langle \Delta E_{\gamma}/E_{\gamma 0} \rangle = [E_{\gamma}(0) - E_{\gamma 0}]/E_{\gamma 0} = 4.004\%
$$
 (18)

for the mean fractional energy shift of the 2796 keV  $\gamma$  rays. For a  $\gamma$ -ray energy of  $E_{\gamma 0}$  = 2795.64  $\pm$  0.7 keV (see Sec. IIIB3 below) we expect, from Eq. (18), a mean full Doppler shift,  $\Delta E_{\nu}$ , of 111.9  $±0.6$  keV. The uncertainty is due almost entirely to that in  $\langle \cos \theta_{n} \rangle$ . The second term on the righthand side in Eq. (16) gives rise to a spread in  $\Delta E$ . which we find is well-simulated by a Gaussian with a FWHM of 4.03 keV. In comparison the spread in ' reaction energies [see Eq. (17)] gives rise to a spread in  $\Delta E_v$  of ~1.5 keV.

#### 2. The measurement

The <sup>4</sup>He-implanted targets were bombarded with  $^{18}$ O ions of 20.1-20.6 MeV and with intensities of 150-300 nA of the 4' charge state. The beam spot was  $\sim$ 2 mm in diameter as defined by a collimator and observed by discoloration of the target after bombardment. The Ge(Li) cylindrical detector of 48.5-mm diameter was placed at  $0^{\circ}$  to the beam with its axis of symmetry on the beam axis and its front face 10.5 cm from the target. A  $^{24}$ Na source was located near the target to provide an accurate energy reference in the Ge(Li) data. The detector efficiency was 16% relative to a  $3 \times 3$ -in. NaI(T1) detector, and was also calibrated independently for  $\gamma$ -ray energies up to 2.614 MeV. To reduce the neutron flux, 6 cm of paraffin were placed between the target and detector. For each set of targets data were recorded using a 4096-channel analyzer for approximately 5 h per target. The net yield of the 2796-keV peak from the second set of targets averaged  $\sim 20\%$  more than that of the first set. A major experimental difficulty was that the  $^{12}C(^{18}O, 2n\gamma)^{28}$ Si (4617 – 1779) reaction initiated on carbon impurities gave a background under the shifted 2796-keV  $\gamma$  ray. To minimize this effect care was taken in the 'He implantation (see Appendix). Also, during the measurements, the vac-



FIG. 4. Portions of  $\gamma$ -ray spectra obtained from 20.1-MeV  $^{18}$ O bombardment of thick Ta with the  $\gamma$ -ray detector at  $0^\circ$  to the beam. The top spectrum was obtained with the <sup>4</sup>He-implanted target and the bottom spectrum from a target consisting of 5  $\mu$ g/cm<sup>2</sup> of carbon evaporated onto a clean Ta blank. The energy dispersion is 0.49 keV/channel. The intensity scales of the two spectra are arbitrary.

uum in the beam line and target chamber was kept at  $10^{-7}$  Torr and the target position was changed to obtain a new beam spot approximately every hour during the measurements. (The average yield for each beam spot served as an internal monitor on the local  ${}^{4}$ He density.) The portion of the  $\gamma$ -ray spectrum displaying the shifted 2796keV transition as obtained for the first Ta target is shown in Fig. 4. Also shown is a spectrum obtained from bombardment of a  $5-\mu g/cm^{2}$  <sup>12</sup>C target evaporated on a clean Ta blank. For each of the targets, the raw spectra were corrected for the <sup>12</sup>C background by subtracting such  $^{18}O+^{12}C$  spectra, suitably normalized. Next, the shape and intensity of the Compton distribution, as determined from the nearby 2754-keV  $^{24}\text{Na}(\beta^-)^{24}\text{Mg}$   $\gamma$  ray, was subtracted. Finally, the remaining background, which displayed no noticeable structure near the  $<sup>21</sup>$ Ne peak, was subtracted assuming a first-order</sup> polynomial shape and using least-squares fitting to the regions just above and just below the  $\mathrm{^{21}Ne}$ peak. Representative spectra for the first set of Mg and Ta targets are shown in Fig. 5.

From the various measurements on the  $^{21}$ Ne  $\gamma$ ray spectra made during this investigation, an accurate energy determination of the  $^{21}$ Ne 2789  $\rightarrow$  0



FIG. 5. The full-energy-loss peak of the  $^{21}$ Ne 2796 $\rightarrow$ 0 transition observed at 0' to the beam after bombardment of  ${}^{4}$ He implanted in Mg (top) and Ta (bottom). The energy dispersion is 0.49 keV/channel. Background and the counts due to the Compton distribution of the 2796-keV  $\gamma$  ray have been subtracted as described in the text. The theoretical line shapes, described in the text, are shown to illustrate the sensitivity of the results to the mean life  $\tau$  of the 2796-keV level.

transition was extracted. The 2789-keV level has a mean life of  $117 \pm 7$  ps<sup>4</sup> and so the 2789-keV  $\gamma$  ray was not Doppler shifted in these measurements. The measurement was relative to the  $^{24}$ Na  $\gamma$  ray of 2754.030  $\pm$  0.014 keV<sup>35</sup> with the <sup>24</sup>Na  $\gamma$  ray of 1368.633 ± 0.006 keV<sup>35</sup> and the  ${}^{1}H(n,\gamma){}^{2}H \gamma$  ray of  $2223.247 \pm 0.016$  keV<sup>36</sup> as secondary standards. The result was  $2788.67 \pm 0.10$  keV corresponding to an excitation energy of  $2788.87 \pm 0.10$  keV.

### 3. Extraction of the mean life

Briefly, the mean life  $\tau$  of the 2796-keV level was extracted from a least-squares fit to the centroid of the 2796-keV line versus the inverse stopping time of  ${}^{21}\text{Ne}$  in the four targets. That is, the attenuation factor  $F(\tau)$  for the *i*th target is defined experimentally by

$$
[F(\tau)]_{i} = [\langle E_{\gamma}^{(i)} \rangle - E_{\gamma 0}] / \Delta E_{\gamma}, \qquad (19)
$$

where  $\langle E_{\nu}^{(i)} \rangle$  is the energy centroid of the line shape distribution for the ith target. Combining Eqs.  $(11)$  and  $(19)$  we have

$$
\langle E_{\gamma}^{(i)} \rangle = E_{\gamma}(0) - \tau \Delta E_{\gamma} [\alpha_c^{(i)}]^{-1}
$$
 (20)

for  $\tau/\alpha_c \ll 1$ . Thus a least-squares fit of a first-



FIG. 6. Peak centroid versus inverse stopping time for the  $^{21}$ Ne 2796-keV full-energy-loss peak observed following  $^{18}$ O bombardment of  $^{4}$ He implanted in Mg, Al, Ta, and Au. The data for the first and second sets of targets are displaced vertically and correspond to the energy scales indicated. The solid lines are least squares fits of Eq. (20) to the experimental points. The values obtained for  $E_{\gamma}(0)$  and  $\tau$  are indicated. These results were obtained neglecting the uncertainty in  $\alpha_c^{-1}$ . The evaluation of the stopping time  $\alpha_c$ , and the effect of this uncertainty, are discussed in the text.

order polynomial to  $\langle E_{\gamma}^{(i)} \rangle$  versus  $[\alpha_c^{(i)}]^{-1}$  will yield an intercept corresponding to the full Doppler shifted energy  $E_{\gamma}(0)$  and a slope of  $-\tau \Delta E_{\gamma}$ . Results of such a fit are illustrated in Fig. 6. The stopping times  $\alpha_c$  were calculated as explained in Sec. II with a correction for the effect of the 'He implantation on  $dv/dt$  as discussed in the Appendix.

The major uncertainty in the centroid energies  $\langle E_{\nu}^{(i)} \rangle$  of Eq. (19) and Fig. 6 arises from the background subtraction and is difficult to evaluate because it is due to the possible systematic deviation of the assumed background from the actual one. The uncertainty was estimated by varying the background subtraction procedures. That is, the relative intensity of the  ${}^{12}C+{}^{18}O$  background subtracted was varied, the remaining structureless background was represented by an exponential as well as a first-order polynomial, and, finally, the energy region included in the least-squares fit to determine the background was varied. The centroids were finally extracted by fitting to the channel region (see Fig. 5) 3408-3448 and the small correction for the omitted high- and lowenergy tails of the line shape was carried out using

the theoretical line shapes. This procedure reduced considerably the sensitivity to the background subtraction and was estimated to introduce negligible error into the mean life extraction. An uncertainty of  $\pm 0.12(0.06)$  keV, corresponding to  $±40(20)$  keV in the mean reaction energy, was added in quadrature to each uncertainty in the mean recoil energy determined in the first (second) set of measurements. The weighted average of the two values of  $E_{\nu}(0)$  and  $\tau$  extracted from the fits illustrated in Fig. 6 were  $2907.54 \pm 0.15$  keV and  $7.9 \pm 0.6$  fs. Using the calculated  $\Delta E$ , of 111.9  $\pm 0.6$  keV we find a value of  $E_{\gamma 0}$  of 2795.64  $\pm 0.7$  keV or an excitation energy of  $2795.84 \pm 0.7$  keV as compared to  $2795.9 \pm 0.6$  keV quoted by Endt and Van der Leun.<sup>5</sup> The uncertainty in the mean life.  $\tau = 7.9 \pm 0.6$  fs, is mainly due to that in the centroid energies and does not include a contribution from  $dE/d(\rho x)$ . Including our estimate of the uncertainty in  $dE/d(\rho x)$ —due almost entirely to the effects of the <sup>4</sup>He implantation (see Appendix)—results in a final value for the mean life of the 2796 keV level of  $7.9 \pm 1.0$  fs. In both the line shape and centroid fitting procedures, second-order effects<sup>12</sup> were taken into account but were found to be negligible. For instance, the correction to  $\tau$  for the  $\beta^2$  term in the Doppler shift amounts to  $2\%$ .

Although the results just quoted could have been obtained without consideration of the  $\gamma$ -ray line shapes, it was felt desirable to investigate these line shapes for two reasons. First, as just discussed, knowledge of the theoretical line shape was helpful in obtaining as accurate energy centroids as possible. Secondly, a successful fitting of the line shapes greatly restricts the possibilities for undiscovered sources of systematic error. Line shapes similar to those of Fig. 3were generated for each target. It was found that for  $\tau$ <10 fs the shape of the  $\gamma$ -ray distribution for Mg was essentially independent of  $\tau$ ; that is, the only measurable effect of changing  $\tau$  was to shift the distribution in energy. Thus, the experimental detector response function used was that of the Mg line shape. This shape had a FWHM consistent with that of the  $^{24}$ Na 2754-keV line convoluted with the calculated kinematical spread of 4.03 keV. Using  $E_v(0)$  = 2907.54 keV and a target density corrected for the  ${}^{4}$ He implantation (see Appendix), the line shapes were calculated as illustrated in Fig. 5. For all targets normalized values of  $\chi^2$  close to unity were obtained for  $\tau \sim 8$  fs. This was indeed reassuring; however, it should be emphasized that no one of the line shape fits by itself would have resulted in a meaningful measurement of  $\tau$ . And, even taken all together, the line shape fits give an accuracy for the determination of  $\tau$  inferior to that in the centroid fitting procedure —as

long as the two procedures are applied independently. If, however,  $E_{\gamma}(0)$  is determined from the centroids as in Fig. 6, then the line shape fits yield an average value for  $\tau$  with comparable accuracy to the centroid procedure.

### C. The 1747-keV level

As shown in Fig. 1, the <sup>21</sup>Ne  $\frac{7}{5}$  + 1747-keV leve decays 95% to the 351-keV first-excited state. The line shape of the 1396-keV  $1747 \div 351$  transition was observed with good statistics, for all four implanted targets, in the same spectra recorded for study of the 2796-keV level. The line shapes were analyzed in a fashion similar to that described for the 2796-keV transition except that the Compton distribution was not subtracted, but was instead included as part of the detector response function. The background was structureless except for the <sup>40</sup>Ar( $\beta$ <sup>-</sup>)<sup>40</sup>K 1461-keV  $\gamma$  ray just above the line shape and the second-escape peak of the <sup>21</sup>Ne 2789  $\div$  0 transition in the center of the line shape. Both were subtracted utilizing their known shapes and normalizing the intensity of the second-



FIG. 7. Line shapes observed for the <sup>21</sup>Ne  $1747 \rightarrow 351$ , 1396-keV transition following  $^{18}$ O bombardment of  $^{4}$ He implanted in Mg, Al, Ta, and Au. The attenuation factor  $F(\tau)$  is shown for each target as is the full Doppler shifted energy obtained from a fit similar to that of Fig. 6. The theoretical fits are described in the text.

escape peak of the 2789-keV transition to that of its full-energy-loss peak. The resulting line shapes for the first set of targets are shown in Fig. 7. There is no feeding from higher-lying levels since the only ones formed —the 2789- and 2796-keV levels —do not decay to the 1747-keV level. The cross section for the 1747-keV level was also found to be resonant at  $E^{(18)}O \approx 19.825$  MeV, and the fractional Doppler shift was also calculated to be 4.004% for the 1396-keV  $\gamma$  ray [see Eq.  $(19)$ ; however, because the reaction is further from threshold than for the 2796-keV level, the kinematical spread is considerably more. It was found to be well simulated by a Gaussian with FWHM= 5.3 keV as compared to the detector response with FWHM= 4.4 keV.

As a first step in the extraction of  $\tau$ , the full Doppler-shifted energy  $E<sub>v</sub>(0)$  was extracted from a fit of Eq. (20) in a manner similar to that illustrated in Fig. 6, with the result  $E<sub>v</sub>(0)= 1452.3$  $\pm 0.1$  keV. Subtracting the expected shift  $\Delta E$ .  $= 55.9 \pm 0.3$  keV, yields  $E_{\gamma 0} = 1396.4 \pm 0.3$  keV as compared to the value of  $1394.83 \pm 0.20$  keV expected from the compiled excitation energies.<sup>5</sup> The agreement is very poor. We note that Grawe The agreement is very poor. We note that Graw<br>et al.<sup>6</sup> obtained  $E_{\gamma 0} = 1396.4 \pm 0.4$  keV for the 1747  $-351$  transition and this value is not included in the set used to obtain the compiled' excitation energies. The excitation energy of the 1747-keV level corresponding to  $E_{\gamma 0} = 1396.4 \pm 0.3$  keV is 1747.2  $\pm 0.3$  keV using 350.72 $\pm 0.06$  keV<sup>5</sup> for the first-excited state.

In the centroid fit to Eq. (21), which yielded  $\tau$  $= 70 \pm 6$  fs, the modification of the stopping times = 70 ± 6 fs, the modification of the stopping times<br> $\alpha_e^{(i)}$  due to the <sup>4</sup>He implantation was estimated only roughly. In addition, the approximation of a constant  $dE/d(\rho x)$  introduces some uncertainty because  $\alpha_e \gg \tau$  is not well satisfied. Thus, the final and best value for  $\tau$  was obtained from the line shape fits shown by the solid curves in Fig. 7. After the preliminary fits to check on the kinematical conditions, the mean life  $\tau$  was obtained from a fit to the slope of the line shape in the energy region below the peak. That is, the fit was confined to the region of channels  $<$  455 shown in Fig. 6. This corresponds to flight times $>130$  fs or distances  $>1.56 \mu m$  which is well beyond the implanted region (see the Appendix). As explained at the end of Sec. II, the result so obtained is insensitive to the detector response function and, most importantly, the  $2N$ e recoils are out of the  $4$ He-implantation region and decaying in the target with well-known density and  $dE/d(\rho x)$ . The final average value<sup>37</sup> for the four backing materials obtained for the mean life of the 1747-keV level was



FIG. 8. Line shape observed for the <sup>21</sup>Ne 2867  $\rightarrow$  1747. 1120-keV transition following  $^{18}$ O bombardment of  $^{4}$ He implanted in Ta. The theoretical fit is described in the text.

# D. The 2867-keV level

The <sup>21</sup>Ne  $\frac{9}{5}$ <sup>+</sup> 2867-keV level decays 62% to the 1747-keV level with  $E_{\gamma 0} = 1120.0 \pm 0.3$  keV (see Fig. 1). This  $\gamma$  ray was not observed with enough intensity for a line shape analysis for  $^{18}$ O bombarding energies below 24 MeV. At energies above ~24 MeV  $\gamma$  rays from <sup>18</sup>O + <sup>24</sup>Mg and <sup>18</sup>O + <sup>27</sup>Al fusion evaporation and transfer reactions became very intense relative to the  $^{18}O+^{4}He$  intensities. Therefore, shapes were obtained only for the Ta and Au targets and because the results for Au had poor statistics only the Ta measurements was analyzed. The line shape for Ta was meaured at  $E^{(18)}O = 27$ MeV and is shown with background subtracted in Fig. 8. The theoretical fit is for  $E_{\gamma}(0) = 1171.1$  keV corresponding to a full Doppler shift of 51.1 keV or 4.56%. As in the case of the 1747-keV level the mean life was obtained from the slope of the line shape in the region where the dependence on kinematics, the detector response, and the effects of the  ${}^{4}$ He implantation are minimal. That is, for the line shape of Fig. 8 the fit was confined to channels 960-997. A result of  $61 \pm 6$  fs was obtained for the 2867-keV level.

### IV. DISCUSSION

### A. Comparison with previous methods

We have presented in this report a method for measuring fast lifetimes based on implanted targets and the high-velocity DSA. The result for the

 $\tau = 74 \pm 6$  fs.

2796-keV level is more accurate than obtained in any previous DSA measurements of  $\tau$ <10 fs lifetimes. The success of this extension of precision DSA results to fast lifetimes is due mainly to the use of implanted targets. Their use allows a wide choice of stopping materials and, by stopping the recoils in the target itself, uncertainties in the target thickness are avoided. These two factorsuse of several stopping materials with a wide range of slowing times and stopping in the target itself were crucial to the accuracy of the method for the fast lifetime. A vexing source of uncertainty is that due to the uncertainties in  $dv/dt$  associated with the  ${}^{4}$ He implantation. It is hoped that future studies of the implantation process can reduce the uncertainty associated with this effect.

For the two slower lifetimes, these two factors are not nearly as important. Here, our accuracy is essentially the same as previous high precision DSA results, e.g., Refs. 21, 32, and 38. Important to the accuracy obtained were the higher statistics made possible because the coincidence technique was avoided. One important contribution we made to the  $\tau = 20-100$  fs region was to avoid the uncertainty due to the effect of  ${}^{4}$ He implantation on the slowing time by fitting to a restricted region of the line shape.

There are two alternative DSA techniques which have successfully yielded fast lifetime measurements and it is instructive to compare them to the present technique.

The method of Branford and Wright. This experimental method<sup>27</sup> uses heavy-ion induced reactions, e.g.,  ${}^{12}C({}^{16}O, \alpha){}^{24}Mg$  and particle- $\gamma$  coincidences. The essence of the technique is to compare the attenuation factor  $F(\tau)$  observed for a thin unbacked target to that observed for a backed target. The analysis is quite similar to that presented in Sec. II and the recoil velocities are comparable to those of the present study. This technique is in some respects more versatile and efficient than the present one. Versatile because most nuclei not too far from stability can be formed in high cross-section heavy-ion fusionevaporation or transfer reactions and efficient because the coincidence technique allows the study of many levels simultaneously. However, the difficulty of fabricating thin targets of known thickness and suitable for heavy-ion bombardment limits the versatility of the method. The sensitivity to the target thickness —as exemplified by the discussion below (Sec. IVB1) of the results of Grawe  $et\ al.<sup>6</sup>$ —and the poor statistics engendered by the coincidence requirement are the major reasons why we feel this technique is intrinsically of poorer accuracy for  $\tau$  <10 fs than the present one. However, when carefully applied it is capable of

nearly comparable accuracy, for instance, Wright, Cooke, Fifield, and Poletti<sup>27</sup> quote  $6.2 \pm 1.6$  fs for the mean life of the <sup>24</sup>Mg 8.11-MeV level obtained in  ${}^{12}C({}^{16}O, \alpha) {}^{24}Mg$ . The technique is complementary to the present one.

 $The method of Anttila and Bister.$  In a series of disternal method of Anttila and Bister. In a series of distributional method of nuclear lifetimes utilizing implanted studies<sup>39,40</sup> of nuclear lifetimes utilizing implante targets and the resonant  $(p, \gamma)$  reaction, Anttila, Hister, and co-workers have developed the lowvelocity DSA to an accuracy comparable to that of velocity DSA to an accuracy comparable to that of the present method. In a typical study,  $^{40}$  a  $^{13}$ C target was made by implanting <sup>13</sup>C in Ta and the  $F(\tau)$ values of various  $14N \gamma$  transitions were measured using the <sup>13</sup>C( $p, \gamma$ )<sup>14</sup>N reaction at the  $E_p = 1.15$ -MeV resonance. For this case  $\beta = 0.36\%$  and  $dv/dt$  is dominated by nuclear scattering. The high accuracy depends on the very large magnitude of  $dv/dt$ encountered for such small velocities and on a careful Monte Carlo treatment of the nuclear scattering. The resultant accuracy is also aided by good kinematical conditions and a clever scheme of experimental conditions. Although the analysis is carried out with great care, the nuclear scattering is not well understood theoretically and there is some possibility of systematic uncertainties larger than those estimated.

### B. Comparison with previous results

The published mean life measurements for the three  $2^{\text{th}}$ Ne levels under study are compared to the present results in Table III. All four previous measurements were via the DSA method. One other result for the 1747-keV level, derived from other result for the 1747-keV level, derived from<br>Coulomb excitation,<sup>5,41</sup> is omitted since the measurement was performed too high above the Coulomb barrier to be reliable.<sup>21</sup>

#### 1. The results 0f Grawe et al.

The only previous mean life determination for the 2796-keV level —the main focus of the present study —was the high-velocity inverse  ${}^{2}H(^{20}Ne, p)^{21}Ne$ study —was the ingn-velocity inverse  $n(x, p)$ <br>reaction study of Grawe et  $al$ .<sup>6</sup> In that measure ment  $\gamma$  rays were observed at 45° and 135° to the  $21$ -MeV  $^{20}$ Ne beam in coincidence with protons observed near  $0^\circ$  to the beam. The attenuation factor  $F(\tau)$  was then obtained as the ratio of the observed Doppler shift to that calculated from the kinematics and detection geometry for  $\tau = 0$ . The target consisted of 50  $\mu$ g/cm<sup>2</sup> of deuterated polyethylene evaporated onto Ta. In our opinion, the experiment and its analysis contain uncertainties of sufficient severity to invalidate the conclusion for the 2796-keV level and to increase somewhat  $(z10\%)$  the uncertainties assigned to the mean lives of the 1747- and 2867-keV levels. The sources of uncertainty in question are the following:

			$\tau$ (fs)			
$E_r$ (keV)	Present	$^{9}$ Be( $^{16}$ O, $\alpha$ ) <sup>a</sup>	$^{18}O(\alpha, n)^6$	$^{18}$ O $(\alpha, n)$ $^{\circ}$	$^{2}$ H( $^{20}$ Ne, p) <sup>d</sup>	
1747	74 $\pm 6$	$150 \pm 130$	$76 \pm 12$ $(76 \pm 20)$	$24 \pm 6$	$85 \pm 7$ $(85 \pm 11)$	
2796	$7.9 \pm 1.0$		$27$	$\cdots$	$10 \pm 4$ (20)	
2867	± 6 61	$110^{+150}_{-70}$	$56 \pm 14$ $(56 \pm 18)$	$27 + 5$	$50 \pm 12$ $(50 \pm 13)$	

TABLE III. Comparison of present and previous lifetime results for  ${}^{21}$ Ne.

<sup>a</sup>Reference 8;  $\beta$  = 2.1, Na(Tl) detectors.

References 9, 10;  $\beta \approx 1.0\%$ . The uncertainties assigned to the results are experiment only. The results in parentheses are our revisions obtained by adding  $15\%$ ,  $10\%$ , and  $10\%$  for  $dE/d(\rho x)$ ,  $\rho$ , and the analysis as explained in the text.

 $^{\circ}$ Reference 11;  $\beta \approx 1.0\%$ . See text.

References 6, 7;  $\beta \approx 4\%$ . The results in parentheses are our revisions as explained in the text.

(1) For  $\beta = 4\%$  the flight time of the <sup>21</sup>Ne recoils through a  $50-\mu g/cm^2$  ( $\rho \approx 1$  g/cm<sup>3</sup>) target is ~40 fs. Thus, for  $\tau = 10$  fs 76% of the recoils decay in the target which has a  $dv/dt$  2.5 times smaller than that of Ta. Thus, not only is the measurable effect,  $1 - F(\tau)$ , commensurately smaller than for decay in pure Ta, but the result is also very sensitive to the time spent in the target. (Incidentally, this difficulty illustrates the main limitation to the accuracy of the technique of Branford and Wright. $27$ ) During the measurement the yield was observed to decrease by a factor of 5, while a subsequent measurement indicated the areal density of the target was the same after the measurement as before, to an accuracy of 10%. This was interpreted to indicate a loss of deuterium only, leaving the stopping power almost unchanged. No uncertainty was explicitly assigned to  $\tau$  from this source. It is our contention, however, that a change in density would almost certainly accompany such a drastic change in composition: Then, because the areal density is essentially unchanged, the thickness and thus the average flight time and the fraction of recoils decaying in the target would be directly affected. This could be a quite large effect because the density of carbon is  $\sim$  twice that of polyethylene. An accounting for the effect could lead to a decrease in the extracted mean life by up to 50%.

(2) The calculated full Doppler shift (FDS) in the (2) The calculated full Doppler shift (FDS)<br>measurement of Grawe *et al.*<sup>6</sup> is of the form

$$
FDScalc = Eyoβ[Q1(45°) cos45°- Q1(135°) cos135°],
$$
 (21)

where the geometrical factors —in square brackets—are displayed explicitly because we wish to discuss their assigned uncertainties. That is, there will be contributing errors to FDS<sub>cale</sub> from

possible deviations from  $45^{\circ}$  and  $135^{\circ}$ , i.e., in  $\cos 45^\circ$  and  $\cos 135^\circ$  and also from uncertainties in the solid-angle attenuation coefficients<sup>34</sup>  $Q_1(45^\circ)$ and  $Q_1(135^\circ)$ . For the 2796-keV level Grawe et al. give FDS $_{\text{calc}}$ = 148.0 ± 0.2 keV or a 0.14% uncertainty. Yet they quote  $Q_1(45^\circ) = 0.957 \pm 0.005$  which gives rise to a 0.26% uncertainty or  $\pm 0.38$  keV all by itself. The contributing uncertainties of cos45', cos135°, and  $Q_1(135°)$  are not given by Grawe et aL, but certainly are not negligible especially in view of the small detector-target distance, 6.2 cm.

For these two reasons we feel that the measure-For these two reasons we feel that the measure-<br>ment of Grawe *et al.*<sup>6</sup> on the mean life of the 2796keV level should be interpreted as an upper limit,  $\tau$  < 20 fs, and that the uncertainties assigned by Grawe et al. to the mean lives of the 1747- and 2867-keV levels are too conservative. We suggest a 10% additional contribution and this is indicated in Table III.

### 2. The results of Bailey et al.

The comparison of Table III indicates quite<br>rongly that the results of Bailey  $et al.^{11}$  are strongly that the results of Bailey  ${et}$   ${al.}^{\text{11}}$  are systematically too low. A clue to the origin of a systematic error in the DSA study of Bailey  $et$   $al.$ is that an attenuation factor  $F(\tau) = 0.046 \pm 0.002$ was observed for the  $^{21}$ Ne 351  $\div$  0 transition yielding  $\tau = 3.6 \pm 0.3$  ps, while the mean life of this state (as reviewed by Millener *et al.*<sup>4</sup>) is  $10.18 \pm 0.16$  ps for which  $F(\tau) \approx 0.017$  would have been expected. One possible explanation of this discrepancy is the presence of voids in the evaporated WQ, target used in these measurements so that the  $2^x$ Ne recoils decay partially in vacuum. If voids were present the relationship between  $\tau$  and the effect of the voids would depend on their nature (size) but would reasonably be expected to be larger for 50-100 fs than for 10 ps.

### 3. The results of Rolfs et al.

The results of Rolfs  ${\it et\ al.}^9$  are a re-analysis The results of ROLLS *et al.* are a re-analysition in the Blaugrund<sup>42</sup> approach, of the original using the Blaugrund<sup>42</sup> approach, of the origina<br>mean life results of Pronko *et al*.<sup>10,43</sup> We have repeated this re-analysis using the Blaugrund approach and find essential agreement with the mean lives quoted by Rolfs et al. (Table III).<sup>44</sup> The uncertainties assigned by Rolfs  $et$  al. (Table III) account only for the experimental errors on the  $F(\tau)$ . The uncertainties in parenthesis in Table III include additional estimates of 15% due to  $dE$  $d(\rho x)$ , 10% due to the uncertain target composition and density, and 10% to the inadequacies of the Blaugrund treatment.

#### C. Comparison with shell-model predictions

The present results are compared to shell-model predictions of Millener<sup>45</sup> in Table IV. This calculation, performed in a SU3 basis, uses an inert

 $s^4 p^{12}$  core and a full  $(sd)^5$  space. Results are shown in Table IV for two effective interactions, Kuo-Brown (KB)<sup>46</sup> and Chung-Wildenthal  $(CW)^{47}$ . The  $B(M1)$  were calculated with the bare nucleon g factors and the  $B(E2)$  using oscillator radial wave functions with the size parameter  $b$  fixed at 1.784 fm. An additional charge  $\epsilon$  such that  $e_{\epsilon}$  $=(1+\epsilon)e$  and  $e_{n}=\epsilon e$  was assumed in the calculations of the  $B(E2)$  values: the two entries for  $B(E2)$  in Table IV are for  $\epsilon = 0.5$  and 0.35, with the latter in parenthesis. Table IV supplements results already given<sup>49</sup> for the Kuo-Brown interaction with  $\epsilon = 0.5$ . The overall agreement between theory and experiment is remarkably good. It is perhaps somewhat better for the Kuo.-Brown interaction than for the Chung-Wildenthal interaction, but the difference is hardly significant. In the Nilsson model, the  ${}^{21}Ne \frac{3}{2}$ <sup>+</sup>,  $\frac{5}{2}$ <sup>+</sup>,  $\frac{7}{2}$ <sup>+</sup>, and  $\frac{9}{2}$ <sup>+</sup> levels are described as members of a  $K_{J}=\frac{3}{2}$  band buil on an intrinsic state with four particles filling the No. 6 orbit and one particle in orbit No. 7. This

TABLE IV. Comparison of experiment to shell-model predictions for electromagnetic transitions between the lowest five even-parity states of  $^{21}$ Ne.

Transition			Theory <sup>b</sup>	Experiment <sup>0</sup>
$J_i^{\pi} \rightarrow J_f^{\pi}$	Quantity <sup>a</sup>	KB <sup>c</sup>	$CW^d$	
$rac{5}{2}$ + $rac{3}{2}$ +	B(M1)	0.084	÷ 0.089	$0.072 \pm 0.001$
	B(E2)	28(20)	27(19)	$24 \qquad \pm \quad 3$
	x(E2/M1)	$+0.074$	$+0.071$	$+0.074 \pm 0.005$
	$\tau$ (ps)	8.7	8.3	$10.18 \pm 0.16$
$rac{7}{2}$ $\rightarrow$ $rac{3}{2}$	B(E2)	12(9)	11(8)	9.7 $\pm$ 1.9
	$\tau$ (ps)	1.2	1.3	1.5 $\pm$ 0.3
$\frac{7}{2}$ + $\rightarrow$ $\frac{5}{2}$ +	B(M1)	0.13	0.18	$0.15 \pm 0.01$
	B(E2)	21(15)	20(15)	11 $\pm$ 3
	x(E2/M1)	$+0.20$	$+0.17$	$+0.14 \pm 0.02$
	$\tau$ (fs)	87	63	78 $\pm$ 6
$\frac{1}{2}$ + $\frac{3}{2}$ +	B(M1)	0.27	0.13	$0.20 \pm 0.014$
	B(E2)	1.3(0.88)	0.57(0.40)	$\cdots$
	x(E2/M1)	$-0.071$	$-0.068$	$\cdots$
	$\tau$ (fs)	5.3	11.3	$\pm$ 1.0 7.9
$\frac{9}{2}$ + $\frac{5}{2}$ +	B(E2)	16(12)	15(11)	15 $\pm$ 1.6
	$\tau$ (fs)	145	155	$160\,$ $\pm 18$
$\frac{9}{2}$ + $\rightarrow$ $\frac{7}{2}$	B(M1)	0.22	0.25	0.23 $\pm 0.02$
	B(E2)	11(8)	11(8)	9 $\pm$ 5
	x(E2/M1)	$+0.09$	$+0.09$	$+0.08$ $\pm 0.02$
	$\tau$ (fs)	102	89	98 $\pm 10$

<sup>a</sup> The transition strengths,  $B(EL)$  and  $B(ML)$ , are in Weisskopf units (Ref. 48), the mixing ratios  $x(L+1/L)$  are dimensionless, and the units for the mean lifetimes,  $\tau$ , are given.  $b$  D. J. Millener, see text. The  $B(E2)$  values are given for effective charge parameters of  $\epsilon$  = 0.5 and 0.35, with the latter in parentheses. The mixing ratios and partial lifetimes were

calculated using the experimental excitation energies and  $\epsilon = 0.5$ .

 $\text{C}$  Using the Kuo-Brown interaction (Ref. 46).

Using the Chung-Wildenthal interaction (Ref. 47).

<sup>e</sup> The mixing ratios are from Ref. 5. The partial mean lives are based on the adopted branching ratios of Fig. 1 (from Ref. 5) and the present total mean lives. The  $B(M1)$  and branching ratios of Fig. 1 (from Ref. 5) and the present total mean lives. The B(M1) and  $B(M2)$  values are calculated from the mixing ratios and the partial mean lives. The  $\frac{1}{2}$  +  $\frac{3}{2}$  $D(uz)$  values are calculated from the mixing ratios and the partial mean fives. The  $\frac{1}{2} + \frac{1}{2}$  branching ratio is assumed to be 100%. For the  $\frac{5}{2} + \frac{3}{2}$  transition the results are as given in Ref. 4.

description is remarkably successful. Thus, the good theoretical description of the electromagnetic transitions connecting these states is not surprising. To a very good approximation, they all have the same basic structure<sup>50</sup> and the M1 and  $E2$ transitions connecting them are essentially determined by gross properties of the nuclear structure and are not sensitive to small changes in the wave functions.

The most sensitive parameter in Table IV is the The most sensitive parameter in Table IV is  $B(M1)$ -value (and thus  $\tau$ ) for the  $\frac{1}{2}$ <sup>+</sup>  $\frac{3}{2}$ <sup>+</sup> transi tion. The measured value lies between that for the Kuo-Brown and Chung-Wildenthal interactions. In a SU3 basis the  $\frac{3}{2}$  state is largely [41] (81) with [41] (62) next in intensity (of order  $10\%)$ ,<sup>4</sup> while for the  $\frac{1}{2}$  state, these intensities are reversed. The spin part of the  $M1$  matrix element, which is intrinsically larger than the orbital part and dom-<br>inates the  $\frac{1}{2}$  +  $\frac{3}{2}$  M1 matrix element is essentially due to the  $(81)$  -  $(81)$  piece. Examination of the contributions to the M1 matrix element reveals that the difference in M1 rates for the KB and CW interactions is due to a large change in the relative interactions is due to a large change in the relation<br>intensity of the  $(81)$  component in the  $\frac{1}{2}^+$  state. It is 12% in intensity using the KB interaction and 5.6% using the CW interaction. The reason for this large difference is not immediately obvious; but since the second  $\frac{1}{2}$ <sup>+</sup> state of  $sd)^5$  is largely (81), it reflects a difference in a mixing of the two lower  $\frac{1}{2}$  states in the two interactions. We note then that the experimental  $B(M1)$  for the  $\frac{1}{2}$ <sup>+</sup> note then that the experimental  $B(M1)$  for the  $\frac{1}{2}$ <br> $\rightarrow \frac{3}{2}$ <sup>+</sup> transition, being senstive to the amount of  $\frac{1}{2}$  transition, being sensitive to the amount of (81) in the  $\frac{1}{2}$ <sup>+</sup> state, can be thought of as determin ing it.

The predicted  $B(E2)$  values tend to give better agreement for  $\epsilon = 0.35$  than for  $\epsilon = 0.5$ . This is in agreement with the omnibus  $sd$  shell comparison agreement with the omnibus sd shell comparison<br>of Schwalm et al.<sup>21</sup> who found that the best overal agreement between the Chung-Wildenthal interaction and experiment for the  $B(E2)$  values of lowlying transitions in the range  $20 \leq A \leq 37$  was obtained with an effective charge parameter of  $x$ called with an effective charge parameter of x<br>= 0.35 using  $b = 1.072 A^{1/6}$  fm (which for  $A = 21$  yields)  $b = 1.781$ .

As stated in the Introduction, the main motive for this study was to provide a value for the  $B(M1)$ of the  $\frac{1}{2}$ <sup>+</sup>  $\rightarrow \frac{3}{2}$ <sup>+</sup> transition accurate enough for use in the evaluation of the to-be-measured parity nonconserving matrix element. A secondary motive was to provide further data for the testing of shell-model calculations so that the predictions' relating to the parity nonconserving matrix element can be made with more confidence. The successful accomplishment of these objectives demanded the further development of the DSA method for use with implanted targets and fast lifetimes. Since a careful examination of all facets of the

measurement and analysis was a necessary part of the attained accuracy, these were described in some detail. The method described should be capable of higher accuracy than presently attained and is applicable to a number of measurable lifetimes in the 1-100-fs range.

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# APPENDIX: 4He-IMPLANTED TARGETS FOR DSA MEASUREMENTS

The utilization of light-ion implanted targets for DSA measurements of nuclear lifetimes has been reported by numerous investigators, and representative examples illustrating the implantation of noble gases in metals of  $A > 48$  are specifically of noble gases in metals of  $A > 48$  are specifically<br>cited.<sup>29-32</sup><sup>39-40</sup><sup>51</sup> Experimental data on the characteristics of these targets —such as the relative concentrations and distributions of the implante<br>ions—have also been presented.<sup>29,39-40,51-55</sup> concentrations and distributions of the in<br>ions—have also been presented.<sup>29,39-40,51</sup>

Most of the precision DSA measurements thus far reported are for relatively longer lifetimes  $(\tau \geq 50 \text{ fs})$ . For such cases the mean decay distance of the recoil excited nucleus is appreciably greater than the spatial thickness of the implanted target layer, and the approximation is therefore quite well justified that the slowing power,  $dv/dt$ , is that of the pure host material. Or, as in the present analysis for the 1747- and 2867-keV levels, the approximation can be avoided by excluding that part of the line shape corresponding to decay in the implanted region. However, the application of the DSA technique to the measurement of shorter nuclear lifetimes (say  $\tau \leq 20$  fs) is not so well understood because of the questions which arise as to the value of  $dv/dt$  appropriate to the implantation region. The nature of the problem can be most easily understood from a preliminary consideration of the present measurements for  $2N$ Ne in the <sup>4</sup>He-implanted targets. For  $\gamma$  emission from a <sup>21</sup>Ne state of  $\tau \sim 8$  fs, the mean decay distance is  $v\tau \approx 0.10 \mu m$ , a distance which is of the order of the spatial thickness  $(0.1-0.4 \mu m)$  of the <sup>4</sup>He distribution in the implanted target metals.

The stopping time  $\alpha_{c}$ , which is the critical energyloss parameter [see Eq.  $(11)$ ], is therefore influenced by the presence of the 'He-implanted atoms and must be taken into account explicitly.

In this Appendix we begin by describing the mechanical details of the target preparation. Experimental measurements of known resonance structure<sup>33</sup> in the <sup>4</sup>He(<sup>18</sup>O,  $n\gamma$ )<sup>21</sup>Ne reaction were employed to deduce the spatial distribution and atomic concentration of the  ${}^{4}$ He implanted in the various host metals. These results were compared and found to be in satisfactory agreement with the <sup>4</sup>He profiles expected from simple calculations based on transport theory. Finally, we have attempted to account, in a quantitative way, for the resultant influence on  $dv/dt$  due to the implantation process.

#### 1. Preparation of 4He-implanted targets

Targets of high purity  $(>99.9\%)$  natural Mg, Al, Ta, and Au were implanted with <sup>4</sup>He<sup>+</sup> ions using a HVEC duoplasmatron ion source operated at extraction voltages providing selected energies  $E^{(4)}$ He)  $\leq 45$  keV. Target blanks of thickness 250- $500 \mu m$  were first etched to remove surface contaminants and then covered by a thin ( $\leq 15 \mu$ g/cm<sup>2</sup>) protective layer of evaporated NaCl. The blanks were clamped to a brass target holder which maximized heat conduction from the implantation area such that the surface temperature did not exceed ~100 °C. The implantations were accomplished by ~1 h bombardments using  $20-30$   $\mu$ A beams of <sup>4</sup>He<sup>+</sup> ions, which were deliberately defocussed over a cross-sectional area of 1.0 cm'. This provided typical implantation doses of  $\sim 6 \times 10^{17}$  He-atoms/ cm<sup>2</sup> corresponding to an areal density of  $-4 \mu g/cm^2$ of  ${}^{4}$ He. The  ${}^{4}$ He bombardment was done in a highvacuum environment ( $< 2 \times 10^{-7}$  Torr) in order to reduce the deposition of contaminant impurities on the target site. After implantation, the NaCl layer was washed away, removing all visible traces of a very slight carbon buildup.<sup>56</sup> traces of a very slight carbon buildup.

Two sets of targets were prepared in this way. The first set was made at a uniform 'He bombarding energy of 40 keV for a net incident bombardment dose ( $\Phi_{inc}$ ) of 6.7 × 10<sup>17</sup> <sup>4</sup>He-atoms/cm<sup>2</sup> for each target. The energy loss in the  $10-\mu g/cm^2$ NaCl layer was  $\neg 6$  keV, so that the <sup>4</sup>He energy incident on the metal surface was actually -34 keV. Because the ranges of  ${}^{4}$ He in different metals vary considerably, this procedure resulted in targets implanted to varying depths, introducing minor but undesirable complications into the measurement and analysis of the DSA data, as discussed in the main text.

At a later date, therefore, a second set of tar-

gets was prepared in which the 'He bombarding energies were calculated to provide implantations to a depth corresponding to a uniform  $^{18}$ O energy loss [for  $E(^{18}O) \sim 20$  MeV] of ~420 keV for all targets. With these targets the  $F(\tau)$  measurements could all be made using nearly the same  $^{18}$ O energy to populate the  $^{22}$ Ne resonant states leading to the  $^{21}$ Ne 2796-keV level. The measurements with this second set of targets provided a more accurate determination of  $\tau$ , in part because the statistics and signal/background ratio were better, and also because the uniform implanted depth reduced the possibilities for systematic error's in the analysis. In the following we therefore describe in detail the analysis to determine the character of the targets used in the second set of measurements, noting that a similar analysis for the first set was also satisfactor ily accomplished.

#### 2. Distribution of the implanted <sup>4</sup>He ions

Pertinent data on the 'He-implanted targets used in the second set of DSA measurements are collected in Table V. For each target the crystalline structure and density are indicated, as well as the <sup>4</sup>He bombarding energies  $[E($ <sup>4</sup>He) and dose  $[\Phi_{\text{inc}}]$  used in the target implantations. The loss in the NaCl protective layer at these 'He energies is ~0.6 keV cm<sup>2</sup>/ $\mu$ g, so that the <sup>4</sup>He energies incident on the metals are lower than the tabulated values of  $E(^{4}He)$  by 6-9 keV.

Yield curves for the  ${}^{4}$ He( ${}^{18}$ O,  $n\gamma$ )<sup>21</sup>Ne reaction were measured at  $\theta_r = 90^\circ$  for each of the eight implanted targets, using a calibrated Ge(Li) detector. Pertinent data for the second set of targets, showing the intensity of 2438-keV  $\gamma$  rays resulting from de-excitation of the <sup>21</sup>Ne  $J^{\dagger} = \frac{1}{2}$  state, are shown as a function of  $^{18}$ O bombarding energy in Fig. 9. The data have been fitted with Gaussian shapes to extract the apparent resonance energies  $(E')$  and resonance widths  $(\Gamma')$ . In order to ascertain the uniformity of the target implantation, the resonant yield of 351-keV  $\gamma$  rays was measured as the <sup>18</sup>O beam was scanned across the diameter of the target spot. The results, shown in the insets, indicate a satisfactorily flat radial density distribution of 'He.

Absolute cross sections for the  $^{18}O(^{4}He, n)^{21}Ne^{*}$ reaction have been measured by Switkowski et  $al.^{33}$ ; these data have been scaled for the inverse reaction  ${}^{4}$ He( ${}^{18}$ O, n) ${}^{21}$ Ne<sup>\*</sup> and are shown in the upper plot of Fig. 9. The data have been fitted with Breit-Wigner shapes to determine the "true" resonance energy  $E_r$  and width  $\Gamma_r$  for the  $J^{\pi} = \frac{1}{2}$  state, as given in Fig. 9.

From the yield curve data for the <sup>4</sup>He-implanted targets, the displacement of the apparent reso-



nance energy  $E'_r$  from  $E_r$  thus provides a direct measure of the mean depth of the <sup>4</sup>He distribution in the host metal. The true resonance width  $\Gamma$ . can then be easily unfolded from the apparent resonance width  $\Gamma'_r$  to yield the width of the <sup>4</sup>He distribution  $\Gamma^{(4}$ He). These values of  $(E'_r - E_r)$  and  $\Gamma^{(4}$ He), given in energy units, were then converted to the corresponding range and distribution parameters  $R_{z}$  and  $\Delta R_{z}$ , using the indicated values of  $dE/d(\rho x)^{18}O$ , as given in Table V. Note that  $R_z$ specifies the mean *projected* range, and  $\Delta R$ , is defined to be the full width at half-maximum (FWHM) of the distribution of ranges.

The integrated absolute yield of the experimental. resonance curves provides a direct measure of the total implanted areal density of  ${}^{4}$ He (defined as  $\Phi$ ) and also the concentration  $C_0$  corresponding to the peak of the <sup>4</sup>He distribution. Here  $C_0$  denotes the percentage of 'He atoms expressed as a fraction of the original (100%) host atoms.

The experimental data of Fig. 9 indicate clearly the optimum  $^{18}$ O bombarding energy for resonant production of the <sup>21</sup>Ne  $J^{\pi} = \frac{1}{2}$  level. The optimum energy for production of the  $J^{\pi} = \frac{1}{2}^+$  state was calculated from these experimental data on  $R<sub>s</sub>$  and  $\Delta R$ , using the observed displacements of the res- $\Delta R_z$ , using the observed displacements of the res-<br>onance structure as observed by Switkowski *et al.*<sup>33</sup> Note that the resonance structure for production of the <sup>21</sup>Ne  $J^{\pi} = \frac{1}{2}^+$  state is well fitted as a doublet, for the parameters given in Fig. 9.

As given in the main text (Sec. III 8) the mean energy and effective width are  $\langle E_{\nu} \rangle$  = 19.825 MeV and  $\langle \Gamma_{\alpha} \rangle$  = 560 keV. The <sup>18</sup>O bombarding energy which leads to maximum production of the  $J^{\pi} = \frac{1}{2}^{+}$ state is then calculable from Table V as  $E_n(^{18}O)$  $= \langle E_{\mu} \rangle + \Delta E$ , where  $\Delta E = R_{\mu} [dE/d(\rho x)]$  is the energy loss of the <sup>18</sup>O beam in arriving at the peak of the 'He distribution. The bombarding energies (in MeV) calculated in this way are Au (20.30), Ta (20.24), Al (20.19), and Mg (20.24). The implantation energies  $E^{(4)}$ He) were initially chosen to allow the same <sup>18</sup>O bombarding energy for all four targets. The observed differences indicate uncertainties due to such problems as surface contaminations, changes during implantation, uncertainties in the predicted  $R_{\rm z}$  for both <sup>4</sup>He and <sup>18</sup>O, etc., and illustrate the desirability of measuring excitation functions, as we have done.

While the energies calculated above provide for maximum yield, it was felt to be more important to require that the mean reaction energy  $\langle E^{(18}O) \rangle$ for production of the <sup>21</sup>Ne  $J^{\pi} = \frac{1}{2}^{+}$  state be the same for all targets: because the mean  $\gamma$ -ray Doppler shift is proportional to  $\langle E^{(18)} \Omega \rangle^{1/2}$  this procedure reduces the possibility for systematic errors in the interpretation of the data. We choose  $\langle E^{(18}O) \rangle$  $= \langle E_{\star} \rangle$ , and the <sup>18</sup>O bombarding energies were de-

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TABLE V.

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FIG. 9. Resonance cross sections (upper plot) for production of the <sup>21</sup>Ne  $J^{\pi} = \frac{1}{2}^{+}$  and  $\frac{1}{2}^{-}$  states via the  $^{4}$ He(<sup>18</sup>O, n)<sup>21</sup>Ne<sup>\*</sup> reaction: from the data on the inverse reaction as given in Ref. 33. The data are fitted with Breit-Wigner shapes for the resonance parameters indicated. The lower plots show  $\gamma$ -ray yield curves measured for production of the  $J^{\pi} = \frac{1}{2}$  state via <sup>18</sup>O bombardment of <sup>4</sup>He-implanted targets. These data were fitted with Gaussian shapes, as shown, to determine the distribution of  ${}^{4}$ He in the implanted target, as characterized by the data on  $R_z$ ,  $\Delta R_z$ ,  $\Phi$ , and  $C_0$  given in Table V. For comparison the mean recoil decay distance  $v \tau$  is shown for  $v/c = 0.04$  and  $\tau = 8$  fs. For each target, the inset shows a scan of the circular target spot along a diameter, illustrating the relative uniformity of the implantation.

termined by folding in the actual  $J^{\pi}$ = $\frac{1}{2}^+$  resonanc structure with the observed 'He distributions. These energies (in MeV) for the four targets are Au (20.60), Ta (20.30), Al (20.25), and Mg (20.30). These latter values are uniformly greater than the first set because of the interplay between the widths of the doublet resonance structure and the variation of 'He distribution widths for the four different targets (see Fig. 9 and Table V).

In Table VI the experimental distribution parameters  $R$ , and  $\Delta R$ , are compared to the values taken eters  $R_z$  and  $\Delta R_z$  are compared to the values take<br>from the compilation of Zeigler,<sup>57</sup> given as  $R'_z$  and  $\Delta R'_s$ . The values of  $R'_s$  correspond to the <sup>4</sup>He energies actually incident on the metal, i.e., after correcting for the energy loss in the NaCl layer (see Table V). Values of  $\Delta R'_g$  correspond to the incident energies,  $E(^{4}He)$  of Table V, because of the expectation that the net straggling in ranges is proportional to the total 'He energy loss.

The predicted values for  $\Delta R'$ , are seen to be somewhat less than the experimental values  $\Delta R_{s}$ by 20-30%. Noting an exception for the Al target, the predicted  $R'_z$  are  $\sim 10\%$  larger than the experimental  $R_z$ , in a direction that could be explained by a small buildup of carbon during the implantation. The reflection coefficients (R, computed from Refs. 58 and 59, are also shown in Table VI; surface corrections have been applied. The expected areal density of 'He is then computed from the  $\Phi_{inc}$  of Table V as  $\Phi' = (1 - \theta)\Phi_{inc}$ .

Comparison of  $\Phi'$  and  $\Phi$ , as given in Table VI, indicates that for both the Au and Al targets only 50-60% of the 'He was actually retained in the metal, while for Ta and Mg the retention is  $~100\%$ .

In the following subsections we wish to estimate the effect which the 'He implantations have on the specific energy loss for  $2N$ Ne ions within the implanted regions. In order to allow for the possibility that the effect may be intermediate between the concentrations corresponding to  $\Phi'$  and  $\Phi$ , we adopt for this purpose the intermediate values  $C_0$  given in Table VI, where the quoted errors on  $C_0$  have been set large enough to encompass areal implantation densities ranging from  $\Phi'$  to  $\Phi$ .

Similar measurements for the first set of targets showed reasonably good agreement between observed and predicted <sup>4</sup>He distributions. However, the Mg target implanted to a dose of  $6.7 \times 10^{17}$  <sup>4</sup>Heatoms/cm<sup>2</sup> exhibited a discoloration over  $\sim$ 20% of the central area: The radial scan of the yield indicated little 'He in this area, and only the peripheral region was used in the DSA measurements. In view' of this observation, the Mg target for the second set was implanted to a lesser areal density, as indicated in Table V, and no such problems were encountered. The range  $R<sub>s</sub>$  for the Al target was markedly less than the prediction from

TABLE VI. Comparison of the implanted <sup>4</sup>He profiles predicted by transport theory to those deduced from experiment.  $R_r$  and  $\Delta R_r$  are the mean range and FWHM of the range distribution and  $\Phi$  is the areal density of the implanted  $H$ . The predicted ranges are from Ref. 54, while the reflection coefficients  $\alpha$  are from Refs. 55–57. For comparison, the mean <sup>21</sup>Ne recoil decay distance is  $v\tau = 0.096 \mu m$ , for  $v/c = 0.04$  and  $\tau = 8$  fs. The values adopted for the peak concentration  $C_0$  are indicated.

Predicted					Experiment				
Host metal	$R_{\star}$ $(\mu m)$	$\Delta R_{\rm g}^{\prime}$ $(\mu m)$	$\Re$ = reflection coeff.	$\Phi'$ <sup>a</sup> $(10^{17} \text{ atom/cm}^2)$	$R_{z}$ $(\mu m)$	$\Delta R_{\star}$ $(\mu m)$	Φ (10 $^{17}$ $atom/cm2$ )	$C_0$ adopted (at, %)	
Au	0.097	0.155	0.15	6.6	0.097	0.230	3.3	$32 \pm 8$	
Ta	0.107	0.126	0.13	7.0	0.094	0.141	6.7	$83 \pm 10$	
Al	0.314	0.178	0.01	6.6	0.200	0.219	3.9	$32 \pm 8$	
Mg	0.385	0.207	0.01	5.2	0.328	0.316	5.4	$44 \pm 8$	

<sup>a</sup> Computed from the  $\Phi_{\text{inc}}$  of Table V as  $\Phi' = (1 - \Re)\Phi_{\text{inc}}$ .

 $\rm{Zeigler,}^{57}$  which may  $\rm{result}$  from an erosion of the surface metal due to the implantation. The second Al target was therefore implanted at a lower <sup>4</sup>He beam current (20  $\mu$ A) for a dose of only  $4.7 \times 10^{17}$  atoms/cm<sup>2</sup>. Although the mean range R<sub>4</sub> agreed more closely with prediction, the retained <sup>4</sup>He was again only  $\sim 50\%$  of the bombardment dose. Accordingly, the second set of DSA measurements employed the Al target made in the first production run, because the yield was a factor of  $\sim 2$ greater.

# 3. Effect of the implantation on  $dv/dt$

The stopping power  $dE/d(\rho x)$  for the implanted areas may be calculated for the atomic concentrations given in Table VI using Bragg's law and tabulated stopping powers for the host material and implanted 'He. However, the quantity that is required for interpretation of the DSA data is the slowing power  $dv/dt$  (or alternatively  $dE/dx$ ) which can be obtained only if we can establish the appropriate density for the implanted region. It is therefore necessary to consider quantitatively the extent to which the implanted <sup>4</sup>He may have deformed (expanded) the lattice structure of the host medium, and perhaps also the form in which it is retained.

There are two main bodies of evidence which may be brought to bear on this question: (i) a quasitheoretical interpretation based on our understanding of the implantation mechanism, and (ii) empirical evidence on stopping powers derived from previous implantation exper iments. The conclusion from these two considerations, discussed separately in the following subsections, is that the stopping power in the implanted region is less than that of the pure host material, in an amount approximately equal to that provided by the expansion due to the implanted He atoms.

The importance of the latter conclusion can be better understood from a simple calculation of the relative volumes of the implanted He and that of the target layer in which it is imbedded. At liquid density  $(0.12 \text{ g/cm}^3)$ , a dosage of  $6.7 \times 10^{17}$  He atoms/cm<sup>2</sup> corresponds to a layer 0.38  $\mu$ m thick: this should be contrasted to the implantation thicknesses  $\Delta R_s$  (0.1–0.3  $\mu$ m) in which the <sup>4</sup>He occupies only a fraction of the total volume.

#### 4. Slowing power: theoretical

It is well known that the solubility of noble gasses in metals is very low —corresponding to only <sup>a</sup> few parts per million (ppm). At concentrations of 100-200 ppm for 'He implanted in W, Picraux and Vook<sup>60</sup> demonstrated via channelling investigations that the <sup>3</sup>He was located substitutionally in clusters of 2 and 3 helium atoms per substitutional site, with clusters of 3 appearing more frequently than clusters of 2. The possibility of clusters involving >3 helium atoms was explored in theoretical cal-<br>culations by Wilson, Baskes, and Bisson.<sup>61</sup> For culations by Wilson, Baskes, and Bisson. For He implanted in an fcc metal (specifically Cu) they found that complexes of 5-8 He atoms in a vacancy could form rapidly at room temperature. The most probable cluster was 6-He atoms per vacancy  $(He_{\alpha}V)$ , and clusters involving fewer He atoms would rapidly coalesce into the  $He<sub>e</sub>V$  configuration. The volume change associated with such clusters, The volume change associated with such cluste<br>as calculated by Baskes and Holbrook,  $62$  corresponds to pressures in the range  $-1.5 \times 10^6$  atm for the  $He<sub>6</sub>V$  configuration.

As the implanted concentrations are raised into the region of a few percent, He bubbles  $15-30$  Å diameter are apparent under examination with diameter are apparent under examination with<br>electron microscopy.<sup>63</sup> However, these bubbles which are several hundred Å apart and correspond to a volume density of  $\sim 10^{15}$  bubbles/cm<sup>3</sup>, contain only a percent or so of the total stored He. Finally, at concentrations greater than 100% (corresponding to doses  $>10^{18}$  He-atoms/cm<sup>2</sup>) the bubbles are observed to coalesce into very large  $(>300 \text{ Å})$ bubbles which eventually rupture through the surface. Investigations of such destructive processes (blistering, exfoliation) suggest a critical saturation whose value is inversely proportional to temperson may, experiencely bugged a critical back<br>tion whose value is inversely proportional to te<br>perature.<sup>64</sup> For the cases studied here it would appear that, for  $T < 200$  °C, this critical concentration would be  $\approx 5 \times 10^{18}$  atoms/cm<sup>2</sup>, i.e., ~7 times greater than that employed for the He-implanted targets.

In the region below this critical concentration, the considerations surveyed suggest that the most likely mechanism for the storage of large amounts of  ${}^{4}$ He involves substitutional clusters of He<sub>V</sub>. with  $n \approx 6$  the dominant configuration. Some fraction of the 'He would undoubtedly be stored interstitially, but the probability for finding individual 'He atoms at substitutional sites should be low, following the expectation that such configurations would be rapidly superseded by configurations of  $n > 1$ .

In the following we examine the volume changes expected for both substitutional and interstitial configurations. Because the theoretical calculations do not include all of the specific cases considered, nor all ranges of concentrations, we quote central values with assigned errors encompassing the range of the calculational variation. As will be seen, this has only a slight effect on the final conclusions.

Interstitial Placement. Calculations for the volume expansion due to interstitial placement of He atoms in bcc metals has been given by Johnson, Bisson, and Wilson<sup>65</sup> in terms of the unit volume  $\Omega$  of the host atoms, from which we adopt a central value  $\Delta V = (0.65 \pm 0.25)\Omega$ . The net fractional volume expansion for a concentration  $C_0$  is thus

 $\Delta V/V = C_0(0.65 \pm 0.25)$ ,

where again  $C_0$  is the peak <sup>4</sup>He concentration.

Substitutional Placement. If the  ${}^{4}$ He is stored in substitutional sites in the form  $He_{\mu}V$ , one then has  $(C_0/n)$  such clusters and correspondingly  $(C_0/n)$ host atoms displaced into interstitial locations. We take  $(0.75 \pm 0.25)$  $\Omega$  as an estimate of the expansion due to interstitial host displacements. For the  $He_nV$  clusters we use the results of Baskes and Holbrook,<sup>62</sup> who conclude that the volume expansion for individual clusters is nearly linear in *n*, being given (for <sup>4</sup>He in Cu) by  $\Delta V$  $=(-0.80+0.60n)\Omega$ . The net fractional volume expansion corresponding to an atomic concentration  $C_0$ , computed as the sum of these terms, is

 $\Delta V/V = C_0(0.60 - 0.05/n)$ .

As can be seen, the result is only slightly sensitive to  $n$ .

The interesting conclusion from this exercise is that the implantation brings about a net volume expansion whose magnitude is nearly independent of the mode of storage. For the following discussion we therefore adopt the average of the above results,

$$
\Delta V / V = C_0 (0.6 \pm 0.2) ,
$$

in order to compute the effect of the implantation on the specific energy loss parameters.

The results of this computation are shown in Table VII. The fractional volume changes  $\Delta V/V$ are specifically for the listed peak concentrations  $C_{0}$ . Within this expanded volume, the presence of the implanted <sup>4</sup>He brings about a change  $\Delta m/m$  in the net mass and a change in the slowing power defined as  $\Delta s/s$ . The resultant fractional change  $\Delta \epsilon / \epsilon$  in the total specific stopping power ( $\epsilon = dE$ )  $dx$ ), or alternatively in the total slowing power  $dv/dt$ , is obtained directly from  $\Delta V/V$  and  $\Delta s/s$ , and does not involve  $\Delta m/m$ . [Conversely,  $dE/d(\rho x)$ ] does depend on the latter term. ]

The reductions in  $dv/dt$  specified by the quoted values of  $\Delta \epsilon / \epsilon$  are appropriate only for the peak concentrations  $C_0$ . Recoiling <sup>21</sup>Ne will of course travel through a range of target material, and thus the average  $C_0$  and  $\langle \Delta \epsilon / \epsilon \rangle$  will be somewhat reduced for practical cases. In the last column of Table VII we show the reduced values of  $\langle \Delta \epsilon / \epsilon \rangle$ calculated for an 8-fs mean life for the experimental conditions employed in the measurement of the  $^{21}$ Ne 2796-keV lifetime. These values were used in conjunction with the  $dE/dx$  for the pure host materials (Tables VII and II) in analyzing the DSA data.

#### 5. Slowing power: experimental

Of the many experiments carried out using implanted targets, almost none provide any direct evidence on the change in slowing power  $dv/dt$  due to the implantation. In several early experiments targets implanted to saturation were employed to measure resonance widths  $(\Gamma)$  or resonance integrals  $\left[\int \sigma(E)dE\right]$  with considerable precision. However, the parameter of interest in these cases is  $dE/d(\rho x)$ , and therefore the effect of a volume expansion would not influence the interpretation of the results or the conclusions. Conversely, earlier DSA measurements involved lifetimes sufficiently long (relative to the slowing down time) that the effects of the implantation would appear only as minor perturbations in the measurement. The expected effects were subsequently allowed

TABLE VII. Summary of changes in specific stopping powers ( $\epsilon \equiv dE/dx$ ) due to implantation of <sup>4</sup>He at the indicated atomic concentrations  $C_0$ . Here  $\Delta V/V$  defines the volume expansion, while  $\Delta m/m$  and  $\Delta s/s$  account for the added mass and slowing down (within this expanded volume) due to the presence of the  ${}^{4}$ He atoms. The last two columns show the peak values and the mean values (appropriate for  $\tau$ =8 fs) of the fractional change in  $\epsilon$ . These data are specifically for <sup>21</sup>Ne ions of velocity  $v/c = 0.04$  corresponding to  $E(^{21}Ne) = 15.9$ MeV.



<sup>a</sup> For the second set of targets.

<sup>b</sup> The uncertainties are propagated from the quoted uncertainties in  $C_0$  with a 30% uncertainty in  $\Delta V/V$  added in quadrature.

for by increasing the quoted uncertainty on the measured values for the lifetime.

Recently, Bister, Anttila, and Keinonen<sup>66</sup> have reported DSA measurements in the <sup>22</sup>Ne( $p, \gamma$ )<sup>23</sup>Na reaction which were utilized to investigate the character of Ne-implanted targets. Ta blanks were implanted to a saturation dose of 6  $\mu$ g/cm<sup>2</sup> of  $2^{2}$ Ne at 80-keV ion energy, and were used to measure the Doppler-shift attenuation of the  $^{23}$ Na 2640-keV  $\gamma$  ray observed at the  $E_{\rho}$ = 1005-keV resonance. Measurements made at the peak of the resonance curve yielded the result  $F(\tau) = (32.5$  $\pm 0.5$ %, while at a somehwat higher bombarding energy (corresponding to the tail of the resonance yield where the  $22$ Ne concentration is markedly less) the results were  $F(\tau) = (30.7 \pm 0.5)\%$ . As noted by the authors, "this indicates higher stopping in that part of the Ta where there are less  $22$ Ne atoms than at the maximum concentration." For comparison, the values of  $\tau$  computed under the assumption that the slowing power is that of pure Ta, are  $103 \pm 5$  and  $112 \pm 5$  fs, respectively. These results of Blister, Anttila, and Keinonen<sup>66</sup> are thus both qualitatively and also quantitatively in accord with the conclusions reached in subsecin accord with the conclusions reached in subsection 4 above. Note that for  $\tau \sim 100 \text{ fs}$ , the  $^{23}\text{Na}$  recoil velocity obtained in the  $(p, \gamma)$  reaction at  $E_p \sim 1$  MeV is yet so low  $-v/c \sim 0.0002$ —that the product  $v\tau \sim 0.07$  µm is again comparable to the spatial thickness of the  $2^{2}$ Ne-implanted region.

From a quite different viewpoint, Eernisse and Picraux<sup>67</sup> have made direct measurements of the volume expansions induced by the implantation of  $4$ He into thin metal plates of Mo, Nb, and Al. The experiment utilized a precision mechanical apparatus to observe the distortion of plates due to implantations in the range of <sup>4</sup>He fluences  $10^{14} - 10^{18}$  $\frac{\text{ions}}{\text{cm}^2}$ , thus providing a direct measure of the volume expansion  $\Delta V/V$ . At low intensities (<10<sup>15</sup>)  $\sim$ ions/cm<sup>2</sup>) the expansion appears linear with  ${}^{4}$ He concentration, while at higher intensities the expansion is less rapid. These results would appear to be in at least semiquantitative agreement with the volume expansions deduced in subsection 4 above.

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