# Subexcitation Positrons

WERNER BRANDT\*

Department of Physics, New York University, New York, and Centre d'Études Nucléaires, Grenoble, France

AND

HOWARD FEIBUS Bell Telephone Laboratories, Whippany, New Jerse v (Received 20 January 1969)

Lifetime spectra of positrons in positronium (Ps)-forming substances can be affected significantly by the addition of small concentrations of impurities with ionization potentials smaller than those of the pure substances. Such impurities permit Ps formation by subexcitation positrons, i.e., by positrons with kinetic energies less than the threshold energy for the formation of Ps in the pure substance. The impurity-induced Ps yield can increase further under the influence of an electric field at field strengths so moderate that the Ps yield in the pure substance remains unaffected. The quantitative aspects of the impurity-induced electric field effect are indicative of the cross sections for elastic scattering and for energy loss of subexcitation positrons in the substance. Therefore, subexcitation positrons permit the experimental study of electronic subexcitation cross sections in a large class of substances.

### INTRODUCTION

~ typically 30 eV to produce an ion pair. This value CHARGED particle moving in a gas spend drops sharply if a small amount of another gas, an "impurity," is added that has an ionization energy less than the lowest excitation level of the pure gas. ' This so-called Jesse effect is caused mainly by energy transfer between excited gas atoms and impurity atoms. A fine structure remains which Platzman' has attributed to the ionization of impurity atoms by subexcitation electrons. By definition, subexcitation electrons have kinetic energies so low that they cannot excite any electronic levels of the molecules of the pure gas.

The slowing down of subexcitation electrons to thermal energies affects many important transient radiation effects in matter. Yet, quantitative information about subexcitation electrons is difficult to obtain and, therefore, virtually absent. Even the Jesse effect, for example, yields only limited information, because energy transfer overshadows the subexcitation processes. By contrast, the processes encountered by subexcitation positrons can influence the annihilation characteristics of the moving particles themselves and can be studied directly. Specifically, the changes in the positron lifetime spectra of positronium (Ps)-forming substances, as induced by the admixture of small concentrations of impurities, are a measure of the energy loss and scattering cross sections in the pure substances. This opens experimental approaches for determining such cross sections. Aside from their inherent interest, cross sections for subexcitation positrons with an energy of a few eV are relevant in the wide context of transient electronic phenomena in matter, because they are indicative of the corresponding cross sections for subexcitation electrons and their trends with the composition and the morphology of the target material.

Subexcitation electrons and positrons lose their energies slowly in collisions with the atoms and molecules of the substance in which they move. Positrons are unique in that they can form Ps in many substances while falling through a characteristic energy range of the substance, the so-called Ore gap. The Ore gap  $E_{O_{\text{re}}}$ ranges between an upper bound  $E<sub>u</sub>$ , which is approximately equal to the internal ionization threshold of the substance  $V_{\text{ion}}$  and a lower bound  $E_l \simeq V_{\text{ion}} - E_{\text{Ore}}$ , where  $E_{\text{Ore}}$  is equal to the Ps ionization energy in the substance. In many substances,  $E_{Ore}$  is close to the Ps ionization energy  $\frac{1}{2}Ry=6.8$  eV. Positrons bound in Ps and free positrons differ in their annihilation characteristics and can be distinguished experimentally.<sup>3</sup>

An impurity with an ionization energy less than the lowest electronic excitation level of the pure substance introduces a subexcitation Ore gap into the system. In Sec. 1, the effects of a subexcitation Ore gap in the positron lifetime spectrum are derived. One finds that the dependence of the spectrum on the impurity concentration yields ratios of the cross sections of interest. These quantities can be separated further, as shown in Sec. 2, if one studies the effect of an external electric field on the Ps yield<sup>4</sup> as a function of the impurity concentration. Through the choice of impurity, this approach opens the possibility for experimental determinations of the cross sections of the pure substance for electronic particles over a range of subexcitation energies.

### 1. Ps YIELD

The influence of a subexcitation Ore gap on the Ps yield of a given substance is derived in this section. The

<sup>~</sup> Research sponsored in part by the U. S. Atomic Support Agency.<br><sup>1</sup> W. P. Jesse and J. Sadauski, Phys. Rev. **100**, 1755 (1955).<br><sup>2</sup> R. L. Platzman, Radiation Re**s. 2,** 1 (1955).

<sup>&</sup>lt;sup>3</sup> Cf. Positron Annihilation, edited by A. T. Stewart and L. Roellig (Academic Press Inc., New York, 1966).<br>
<sup>4</sup> W. Brandt and H. Feibus, Phys. Rev. 174, 454 (1968).

POSITRONIUM FORMATION IN IMPURITY GAS  $J^{\theta}E_{\text{Ore}}$ ORE I HORE SUBE XCITATION POSITRONS E  $\circ$  $E_z$   $E_u$   $E_d$   $E_u$ 

FIG. 1. Positrons that fall into the Ore gap of a gas,  ${}^gE_{{\rm Ore}}$ , can form positronium (Ps). Subexcitation positrons with energie<br>below  ${}^{\rho}E_l$  annihilate as free positrons. If an impurity is added with an ionization potential  $V_{\text{ion}} < V_{\text{ion}}$ , subexcitation positrons can<br>form Ps in the impurity Ore gap,  ${}^t_0C_{\text{ore}}$ . The energy range between<br>the Ore gaps vanishes if  ${}^t_0V_{\text{ion}} > {}^{\rho}E_t$ , in which case  ${}^t_0E_u = {}^{\rho$ the rate of energy loss of the subexcitation positrons in  $E_{O_{\text{re}}}$ .

treatment applies to Ps-forming substances generally. For definiteness of terminology, the problem is formulated here in terms of a (pure) gas, superscript g, with an impurity, superscript  $i$ , at a concentration (molar fraction)  $c$ . The mixture is referred to as the system, superscript s. The results can be transcribed with only minor modifications to apply to condensed systems. Rates are denoted by  $\gamma$  and cross sections by  $\sigma$ . They are interrelated as  $\gamma = n v \sigma$ , where *n* is the density of scatterers and  $v$  the positron velocity. In most cases, it suffices to set  $v=v_k$ , the positron velocity corresponding to the lower bound  $E_l$  of the relevant Ore gap.

Figure 1 shows schematically the Ore gap of the gas,  ${}^{\rho}E_{\text{Ore}}$ , and the subexcitation Ore gap of an impurity,  $E_{0}$ . After their last ionizing collision with the gas atoms during the slowing-down process, the positrons fall, at time  $t=0$ , into the energy range between zero and  ${}^{\circ}E_u$  with a distribution  $g(E_0)$  of initial energies  $E_0$ , normalized such that

$$
\int_0^{\sigma E_u} g(E_0) dE_0 = 1.
$$

At later times, the energy has dropped to some  $E(t, E_0)$  $\langle E_0$ . Over the limited range of an Ore gap we can set

$$
-dE(t, E_0)/dt = \gamma_r E(t, E_0) = \Lambda \gamma_{sc} E(t, E_0) , \qquad (1)
$$

where  $\gamma_r$  is the mean energy-loss rate characteristic of the energies of the Ore gap,  $\Lambda$  is the fractional energy loss per scattering collision, and  $\gamma_{\rm so}$  is the scattering rate. In most Ps-forming substances the Ps-formation rate  $\gamma_f$  is so large compared to  $\gamma_r$ , that the Ps yield is given by

$$
\sigma \Phi = \int_{{}^{\sigma}E \text{Ore}} g(E_0) dE_0. \tag{2}
$$

Otherwise, Eq. (16) of Ref. 4 applies.

If an impurity with  ${}^{i}V_{\text{ion}} < {}^{g}V_{\text{ion}}$  is added to the gas, some of the subexcitation positrons with initial energies  $E_0 \lt^{\rho} E_i$  form Ps in the impurity Ore gap  $E_{0r}$ . At small impurity concentrations the total Ps yield in the system becomes

$$
{}^s\Phi(c) = {}^g\Phi + {}^i\varphi(c). \tag{3}
$$

The following parameters determine the impurity contribution ' $\varphi(c)$ . The Ps-formation rate in ' $E_{0r}$  is ' $\gamma_f(c)$  $=cn^{i}v_{i}\sigma_{f}$ , while in  ${}^{g}E_{\text{Ore}}$  the formation rate  ${}^{g}\gamma_{f}(c)$  $= (1-c)n^{\rho}v_l^{\rho}\sigma_f \simeq \rho_{f}(0)$  remains essentially concentration-independent. Also, the annihilation rate of the positrons in  ${}^{i}E_{0r}$  remains characteristic of the gas in the energy range of the impurity Ore gap. The probability per unit time of Ps formation in  $E_{O_{\text{re}}}$  becomes

$$
{}^{i}q_{f} = {}^{i}\gamma_{f} \exp[-({}^{i}\gamma_{f} + {}^{g}\gamma_{a})t] \cdot E_{I} \leq E \leq {}^{i}E_{u}, \qquad (4)
$$

and the probability of energy losses in ionizations of impurity atoms between the Ore gaps, where  $i_{\gamma} = 0$ , becomes

$$
{}^{i}q_{\text{ion}} = {}^{i}\gamma_{\text{ion}} \exp[-({}^{i}\gamma_{\text{ion}} + {}^{g}\gamma_{a})t] \cdot E_{u} \leq \mathbf{g} \leq {}^{g}E_{l}. \quad (5)
$$

Following the method developed in Ref. 4, we calculate  ${}^{i} \varphi(c)$  from the Boltzmann equation in momentum space for the positrons in the system. Kith some minor simplifications, the result can be written in the form

$$
{}^{i}\varphi(c) = \int_{{}^{i}E \text{Ore}} dE_{0}g(E_{0}) \int_{E(t, E_{0}) \geq {}^{i}E_{l}} dt^{i}q_{f}(t, c)
$$
  
+ 
$$
\int_{{}^{i}E_{u} \leq E_{0} \leq {}^{i}E_{l}} dE_{0}g(E_{0}) \left[1 - \int_{E(t, E_{0}) \geq {}^{i}E_{u}} dt^{i}q_{\text{ion}}(t, c)\right]
$$
  

$$
\times \int_{E(t, {}^{i}E_{u}) \geq {}^{i}E_{l}} dt^{i}q_{f}(t, c). \quad (6)
$$

The first term is the contribution from the positrons initially in  $E_{O_{\text{re}}}$ . The second term gives the contribution of the positrons between the two Ore gaps. By Eq.  $(2)$ almost all positrons initially in  ${}^{\rho}E_{\mathrm{Ore}}$  form Ps. Therefore, we set  ${}^{i}E_{u} = {}^{g}E_{l}$ , irrespective of  ${}^{i}V_{ion}$ , whenever the two Ore gaps overlap, i.e., for all impurities with  $V_{\text{ion}} > {}^{\rho}E_l$ . The second term in Eq. (6) is then zero. It is generally small at impurity concentrations where Ps formation in  $E<sub>0re</sub>$  becomes noticeable because then  $\gamma_{\rm ion} \gg \gamma_{\gamma}$ . In the following, we neglect this term and, moreover, assume that  $\gamma_r \gg \gamma_a$ . Integration yields

$$
{}^{\circ}\Phi(c) = {}^{\circ}\Phi + \epsilon (1 - {}^{\circ}\Phi) \left[ 1 - (1 - {}^{\circ}\Phi) \frac{{}^{\circ}\lambda_1(c){}^i N(c)}{{}^i\lambda_1(c) - 1} \right], \quad (7)
$$

FIG. 2. Effect of an impurity in a gas on the Ps yield of the system. The abscissa is proportional to the impurity concentration  $c$ . The curves are calculated for the situation shown in Fig. 1. It resembles the system He with Ar as impurity, which is somewhat extreme with regard to the spacing between the<br>Ore gaps. The solid curve shows Eq.  $(7)$ , which is calculated under the assumption that the contribution from positrons between the<br>Ore gaps is small, and that the positron annihilation rate is very small compared to the energy-loss rate. If this is not so, the Ps yield, by Eq. (6), rises somewhat differently as indicated by the broken curves.



where  $\Phi$  is given by Eq. (2),

$$
{}^{i}\Phi = 1 - ({}^{i}E_{l}/\epsilon^{q}E_{l}),
$$
  
\n
$$
({}^{i}E_{n}/{}^{g}E_{l}, \text{ for } {}^{i}E_{n} \leq {}^{g}E_{l},
$$
\n
$$
({}^{i}E_{n}/{}^{g}E_{l}, \text{ for } {}^{i}E_{n} \leq {}^{g}E_{l},
$$
\n
$$
({}^{g})
$$

$$
\epsilon = \begin{cases}\n\epsilon & \text{if } i = 0, \\
1, & \text{for } iE_u > {}^g E_i,\n\end{cases} \tag{9}
$$

and

$$
{}^{i}N(c) = 1 - (1 - {}^{i}\Phi)^{i}\lambda_1(c) / (1 - {}^{i}\Phi)^{i}\lambda_1(c). \tag{10}
$$

The concentration dependence appears through the parameter

$$
{}^{i}\lambda_{1}(c) \equiv c \, {}^{i}\sigma_{f} / {}^{g}\sigma_{r}.
$$
 (11)

Equation (7) expresses the fact that the Ps yield of the system is equal to the yield in the gas plus the fraction of positrons falling into the impurity Ore gap below  ${}^{\rho}E_l$ , multiplied by the concentration-dependent probability of forming Ps there. The illustrative examples in Fig. 2 show that the contribution of the impurity to the total Ps yield is significant even at very small concentrations, because typically  $i_{\sigma} / \sqrt[q]{\sigma} \approx 10^{3}$ -10<sup>4</sup>. At larger concentrations a plateau is reached which is determined by the magnitude of ' $\Phi$ . At still higher concentrations,  $\mathscr{I}\Phi$  does not remain independent of  $c$ , but begins to decrease by a factor  $\sim \frac{\varphi}{f}$   $\left[\frac{\varphi}{f} + \frac{i}{\gamma} \sin(c)\right]$  as ionizations of the impurity atoms in  ${}^gE_{0re}$  begin to compete with Ps formation. The total yield  $\Phi(c)$  can go through a maximum or rise monotonically toward the Ps yield in the pure second component. Such trends have been observed by Deutsch and his co-workers<sup>5</sup> on krypton and freon in argon in the gas phase, and by Paul<sup>6</sup> on

propane in argon in the gas and in the liquid phase. An impurity with an Ore gap higher than that of the gas affects the Ps yield only at large concentrations because  $i_{\sigma}$  must compete with the inelastic processes in the gas. In the following, we shall not be concerned with large impurity concentrations.

In summary, the concentration dependence of the Ps yield of a system with a subexcitation impurity Ore gap measures, through the parameter  $i_{\lambda_1}(c)$ , Eq. (11), the ratio of the Ps-formation cross section of the impurity,  $i_{\sigma f}$ , and of the cross section for the energy loss of subexcitation positrons in the gas,  $\sigma_r$ . To separate the cross sections further, the method must incorporate at least one more experimental variable. The change of the impurity concentration dependence of  $\Phi$ , as induced by an electric field, satisfies this requirement.

### 2. EFFECT OF ELECTRIC FIELD

An electric field  $E$  affects the Ps yield of a gas in two ways.<sup>4</sup> (1) It induces the diffusion of positrons out of the

Kr in Ar at  $c > 10^{-3}$ . In the liquid Ar-propane system the Ps yield at the interest with propane concentration in the very concentration range where no change is observed in the gas phase. Actually, cording to Eq. (7) and Fig. 2, this implies that at a given impurity<br>concentration  $({}^{\tau}\sigma_f/\sigma_{\sigma_r})_{\text{cond}}/({}^{\tau}\sigma_f/\sigma_{\tau})_{\text{gas}} \ll 1$ ; from the data one<br>estimates this ratio to be  $\sim 10^{-2}$  or less. Presumably  ${}^{\tau}\sigma_f$  and are not affected grossly by the condensation of this system, and the change must be attributed to a ratio  $\Lambda_{\rm gas}/\Lambda_{\rm cond}\sim 10^{-2}$  of the fractional energy losses per scattering collision defined by Eq. (1). One should expect such a ratio because the positron energy loss in the gas phase occurs primarily through momentum transfer in elastic collisions with the gas atoms, for which  $\Lambda_{\rm gas} = 2m_+/M(\rm Ar)$  $=3\times10^{-6}$ , while in condensed systems energy loss can occur more<br>efficiently via phonon-type excitations, for which  $\Lambda_{cond} = 2m_+/M_{ph}(\text{Ar})\sim [4m_+c^2/(1-\sigma\Phi)\phi V_{ion}]^{1/2}=2\times10^{-3}$  (c= velocity of sound). (See Ref. 4.) Such a of subexcitation positrons on condensation would affect both  $\Phi$  [cf. Ref. 4, Eq. (16)] and  $\Phi(c)$ , Eq. (7), in just the way indicated by Paul's observations on pure argon and on argon-propane mixtures.

<sup>&</sup>lt;sup>5</sup> M. Deutsch (private communication); E. P. Dulit, B. Gittel-

man, and M. Deutsch, Bull. Am. Phys. Soc. 1, 69 (1956).<br> **CALCE 1998** PD. A. L. Paul; cf. C. Y. Leung and D. A. L. Paul, Ref. 3, p. 409 and references cited therein. Paul investigated the range  $c > 10^{-3}$  and observed the plateau and the high-concentration decline in the Ps yield of gaseous Ar with propane, which is expected to follow the rise at smaller concentrations. The observations are consistent with the data of Deutsch  $el$  al. (Ref. 5) for



 $\epsilon$ /P(V/cm atm)

FIG. 3. The probability  $W(c, E)$  that positrons initially below the impurity Ore gap can form Ps. The curves are calculated for a system similar to Ar impurities in He. The concentration dependence shows an inversion at  $c\simeq 10^{-3}$  for reasons discussed in the text.

Ore gap; this decreases  $\Phi$ . (2) It raises the mean temperature of the positrons below the Ore gap such that the tail of the energy distribution reaches into the Ore gap; this increases  $\Phi$ . Process (1) dominates at moderate fields. Process (2) takes over at high fields, and  $\Phi$ asymptotically reaches a saturation value when practically all positrons form Ps. In many gases the Psformation rate  $\gamma_f$  is so large compared to the energyloss rate  $\gamma_r$ , that the decrease (1) is not observed. For simplicity and without loss of generality in the subsequent discussion, we shall assume this to be the case for the (pure) gas.

Both field-induced processes can occur at the subexcitation energies of the impurity Ore gap and afFect  $\Phi$ . In the notation of Ref. 4,

$$
{}^{\circ}\Phi(c, \mathbf{E}) = {}^{\rho}\Phi + {}^{\dot{\phi}}\varphi(c, \mathbf{E}) + [1 - {}^{\rho}\Phi - {}^{\dot{\phi}}\varphi(c, \mathbf{E})] {}^{\dot{\phi}}W(c, \mathbf{E}), \tag{12}
$$

where  $\mathscr{I}\Phi$  is given by Eq. (16), Ref. 4, or by Eq. (2). The impurity yield  ${}^{i}\varphi(c, E)$  depends on the field as<sup>4</sup>

$$
{}^{i}\varphi(c,\mathbf{E}) = {}^{i}\varphi(c,0) - A(c){}^{i}\rho(c,\mathbf{E}), \qquad (13)
$$

where

$$
A(c) = \epsilon (1 - \sigma \Phi)(1 - \Phi) \left( \frac{3^i N(c)}{2 \left[ \Lambda_1(c) - 1 \right]} \right)^{1/2}, \quad (14)
$$

in terms of the dimensionless variable

$$
{}^{i}\rho(c,E) = (\frac{2}{3}e^{2}/m_{+}{}^{s}\gamma_{r}{}^{s}\gamma_{sc}{}^{i}E_{l})^{1/2}E, \qquad (15)
$$

which is linear in the field strength **E**. The function  $W(c, E)$  in Eq. (12) is the probability that positrons initially below  $E_l$  can form Ps because of the heating of the subexcitation positrons by the field. In the approximation discussed in Ref. 4, we have

$$
^{\bullet}W = (^{\mathbf{i}}\Gamma_{l} - ^{\mathbf{i}}\Gamma_{u} + ^{\mathbf{\sigma}}\Gamma)/(^{\mathbf{i}}\Gamma_{l} - ^{\mathbf{i}}\Gamma_{u} + ^{\mathbf{\sigma}}\Gamma + 1). \quad (16)
$$

The function

$$
{}^{\sigma}\Gamma(E) = \frac{{}^{\sigma}W(0,E)}{1 - {}^{\sigma}W(0,E)} = \frac{{}^{\sigma}\Phi(E) - {}^{\sigma}\Phi(0)}{1 - {}^{\sigma}\Phi(0)} \tag{17}
$$

is known from the Geld dependence of the Ps yield in the pure gas. In terms of Eq. (15), we have

$$
\mathbf{^{\dot{a}}}\Gamma_{l} = \lambda_{2} \big[1 - \left(2/\sqrt{\pi^{i}\rho}\right) \exp\left(-\,^{i}\rho^{-2}\right) - \text{erf}\left(\,^{i}\rho^{-1}\right)\big],\tag{18}
$$

where

$$
i\lambda_2(c) \equiv c^i \sigma_f /^g \sigma_a. \tag{19}
$$

 ${}^{\mathbf{i}}\Gamma_{\mathbf{u}}$  is obtained by replacing, in Eq. (18), the variable  $i_{\rho}$  by  $i_{\rho}(1-i\Phi)^{1/2}$ .

At moderate fields,  $\mathbf{F}(\mathbf{E})=0$  and  $\mathbf{F}_l \gg \mathbf{F}_u$ , so that the field dependence is described by

$$
{}^{\bullet}W(c,\mathbf{E})\simeq {}^{\bullet}\Gamma_l(c,\mathbf{E})/{}^{\bullet}\Gamma_l(c,\mathbf{E})+1\,,\tag{20}
$$

i.e., the entire rise of  $\Phi$  stems from the impurity

Figure 3 shows  $W(c, E)$  for the system specified schematically in Fig. 1. It resembles He with Ar as impurity. The magnitude and velocity dependence of the relevant cross sections are taken from Teutsch and Hughes,<sup>7</sup> with the additional assumption that for Ar,  ${}^{i}\sigma_{1} \simeq {}^{i}\sigma_{\text{sc}}$ . Even at small impurity concentrations, 'W rises significantly for values of  $E$  at which the yield in the pure gas is unafFected. The total field dependence of  $\Phi$ , Eq. (12), is displayed in Fig. 4. Given such curves, the subexcitation cross sections can be extracted as indicated in Ref. 4.

The concentration dependence of the field effect is conveniently discussed in terms of the 6eld strength at which  $W$  has reached a certain value, for example, in terms of  $\mathbf{E}=\mathbf{E}_{1/2}$ , where  $\mathbf{W}(c, \mathbf{E}_{1/2})=\frac{1}{2}$ . While  $\mathbf{\hat{v}}_2(c)$ [Eq. (19)], increases linearly with c, the variable  $\phi$  $[Eq. (15)]$  increases or decreases approximately as



FIG. 4. The reduced Ps yield  $\Phi(c,E)/\Phi(c,0)$  can show an initial decline before rising at field strengths below those necessary to affect the yield in the pure gas. The curves are calculated for a system resembling He with Ar as impurity.

<sup>7</sup> W. B. Teutsch and V. W. Hughes, Phys. Rev. 103, 1266  $(1956).$ 

 $\mathbf{v}_p(c) \simeq \mathbf{v}_p(0)/(1-c^i s)$ , where  $\mathbf{v}_s = 1-(\mathbf{v}_{\rm sc}/\mathbf{v}_{\rm sc})$ , depending on whether 's is positive or negative. On balance,  $\mathbf{E}_{1/2}(c)$ goes through a minimum if  $\dot{s}<0$  (cf. Figs. 3 and 4), but diminishes monotonically if  $i > 0$ : The qualitative behavior of  $\mathbf{E}_{1/2}(c)$  reflects the relative magnitude of  ${}^{\boldsymbol{i}}\sigma_{\rm sc}$  and  ${}^{\boldsymbol{g}}\sigma_{\rm sc}.$ 

In conclusion, small concentrations of impurities can induce Ps formation by subexcitation positrons and thereby change the Ps yield of a substance significantly. At a given impurity concentration, the increase of the Ps yield depends on the ratio of the Ps-formation cross section of the impurity and the energy-loss cross section of the subexcitation positrons [cf. Eq.  $(7)$  and Fig. 2j. The further rise in the impurity Ps yield under the influence of an electric field is regulated, in addition, by the elastic scattering cross section of subexcitation positrons  $\lceil$  cf. Eqs. (12) and (15)]. The energy dependence of the cross sections can be studied to some extent through the choice of impurity. The expected magnitude of the impurity-induced changes in the positron lifetime spectra suggests that they can give experimental access in Ps-forming substances to scattering and energy-loss rates relevant for an understanding of electronic subexcitation processes in matter.

PHYSICAL REVIEW VOLUME 184, NUMBER 2 10 AUGUST 1969

# Selective Excitation of Nuclear Sublevels\*

NEIL D. HEIMAN AND J. C. WALKER Physics Department, The Johns Hopkins University, Baltimore, Maryland 21Z18

AND

LOREN PFEIFFFR Bell Telephone Laboratories, Murray Hill, New Jersey and Physics Department, The Johns Hopkins University, Baltimore, Maryland Z1Z18' (Received 27 January 1969; revised manuscript received 21 May 1969)

A method is described for selectively populating any one of the sublevels of a nuclear excited state by means of the Mössbauer effect. Specifically, a single hyperfine level of the 14.4-keV state of Fe<sup>57</sup> was populated using a constant-velocity drive system, a single-line Co<sup>57</sup> source, and a metallic Fe<sup>57</sup> scatterer arranged in 90' scattering geometry. Polarization was detected by analyzing the scattered radiation with a second Mossbauer drive and a single-line absorber. Two applications of the selective excitation process are also reported. In the first, the energy distribution of the scattered radiation was studied as a function of the Doppler shift of the incident radiation. In the second, NMR transitions between the hyperfine sublevels of the 100-nsec state of  $Fe<sup>57</sup>$  were directly observed.

'HIS paper reports the development of a technique for producing and detecting excited-state nuclear polarization by means of nuclear resonance radiation (the Mössbauer effect). This is the first successful extension to nuclei of the concept of selective excitation of magnetic sublevels that was the basis of numerous experiments using atomic levels.<sup>1</sup> Two applications of the technique are also reported here. In the first, the energy distribution of the resonantly scattered radiation was measured as a function of the Doppler shift of the incident  $\gamma$  rays. In the second, NMR transitions in the 14.4-keV state of Fe<sup>57</sup> were directly observed for the first time. This second application demonstrates that the problems associated with magnetically induced rf sidebands,<sup>2</sup> which completely obscured the results of earlier resonance experiments,<sup>3,4</sup> have been overcome,

that the general technique of Mössbauer-effect detection of excited-state NMR is feasible, and that therefore investigations into the effects of resonant transitions upon the electromagnetic line shape can be performed. The results reported here indicate that such effects, if they exist at all, are smaller than predicted either by Hack and Hammermesh<sup>5</sup> or by Gabriel.<sup>6</sup>

The basic idea of the polarization process is as follows. A constant-velocity transducer  $(CVT)$  and a constant-acceleration transducer are arranged in a right angle with a scattering foil at the apex (see Fig. 1).The CVT supplies the proper velocity to a monochromatic source in order to Doppler-shift the recoillessly emitted  $\gamma$  rays into resonance with a transition to only one of the resolved hyperfine levels of nuclei in the scattering

<sup>\*</sup>Work supported by the U. S. Atomic Energy Commission. '

<sup>&</sup>lt;sup>1</sup> For a survey of such experiments, see, for example, A. Kastler, Physics Today 21, 34 (1967).<br><sup>2</sup> N. Heiman *et al.*, Phys. Rev. Letters 21, 93 (1968); J. Appl.<br><sup>2</sup> N. Heiman *et al.*, Phys. Rev. Letters 21, 93 (1968);

Lustig [University of Illinois Report No. AFOSR TN 60-698<br>(unpublished)]; Phys. Rev. 172, 319 (1968).

<sup>4</sup> E. Matthias, in Hyper6ne Interactions and Nuclear Radiations, edited by E. Matthias and D. A. Shirley (North-Holland Publishing Co., Amsterdam, 1968).

<sup>&</sup>lt;sup>6</sup> M. N. Hack and M. Hammermesh, Nuovo Cimento 19, 546  $(1961)$ 

Helmut Gabriel, this issue, Phys. Rev. 184, 359 (1969).