

"spin" Green's function

$$G_{m_1 m_2; m_3 m_4}(u) = \langle T \{ c_{m_1}^\dagger(u) c_{m_2}(u) c_{m_3}^\dagger c_{m_4} \} \rangle. \quad (27)$$

This Green's function can be evaluated diagrammatically following the methods used by Hernández and Walker.<sup>10</sup> The results can be cast into the familiar form<sup>7</sup> of an equation of motion for the reduced density matrix  $\hat{\sigma}$  which, in the notation of Redfield,<sup>7</sup> is

$$d\hat{\sigma}/dt = i[\hat{\sigma}, \hat{H}_{\text{eff}}] + R\hat{\sigma}. \quad (28)$$

It is clear from Eq. (28) that the energy levels determined in an EPR experiment are the eigenvalues of  $\hat{H}_{\text{eff}}$ . This justifies our choice of definition of  $\hat{H}_{\text{eff}}$ .

The relaxation matrix,  $R$ , is as given by Eq. (3.15) of Redfield's paper,<sup>7</sup> except that the matrix elements appearing there should be replaced by the renormalized effective  $t$ -matrix elements defined by Eq. (22) above.

Strains must be included to obtain agreement with experiment<sup>1,2</sup>; these are included in our theory, following Ham,<sup>1,2</sup> by adding the appropriate term to  $\hat{H}_{\text{eff}}$ . When this is done, Eq. (28) gives a solution to the motional narrowing problem similar to that discussed by Watkins and Ham.<sup>8</sup>

The above techniques allow a rigorous perturbation discussion of the dynamic Jahn-Teller effect in the case of weak coupling to the phonon continuum. Symmetry is properly accounted for, explicit expressions for the important parameters of the theory (i.e.,  $p$  and  $q$ ) are obtained,

and contact is made with experiment through a study of the appropriate Kubo susceptibility. The method should prove useful in the study of other Jahn-Teller problems, e.g., the orbital triplet state, the cooperative Jahn-Teller effect, phonon-induced interactions between pairs of Jahn-Teller ions, and theories of acoustic and optical experiments.

\*Research supported by the National Research Council of Canada.

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## Positron Diffusion in Metals\*

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(Received 11 June 1973)

From positron lifetime measurements in seven powders of Fe, Co, Ni, and W, the positron diffusion constant in metals at 300°K is found to be  $D_+ = (1.0 \pm 0.5) \times 10^{-2}$  cm<sup>2</sup>/sec. This agrees with a theory based on positron-electron scattering. Evidence is presented that positrons are trapped in metal surface states.

The diffusion constant for positrons,  $D_+$ , in metals is the link between absolute determinations of metal-vacancy concentrations  $C$  and mea-

surements of positron annihilation characteristics in the following sense. If vacancies in solids capture thermalized positrons, and if the capture

rate  $\kappa$  is diffusion limited,  $\kappa$  may be written in the form<sup>1,2</sup>

$$\kappa = 4\pi r_v D_+ Cn, \quad (1)$$

where  $r_v$  is the vacancy capture radius ( $\sim 1$  atomic cell radius) and  $n$  is the atomic density. Binding energies of positrons in metal vacancies are of the order of 1 eV.<sup>3</sup> The escape rate from the vacancies in general may be neglected compared to the positron annihilation rate in the vacancy. If two positron lifetime components cannot be resolved, trapping in defects manifests itself in a mean lifetime  $\bar{\tau}$  intermediate between the lifetime in the crystal bulk,  $\tau_c$ , and in the defect,  $\tau_d$ . It depends on  $\kappa$  as

$$\kappa\tau_c = (\bar{\tau} - \tau_c)/(\tau_d - \bar{\tau}). \quad (2)$$

A narrowing of the angular correlation between the two 0.5-MeV  $\gamma$  rays that emerge from the solid when a positron-electron pair annihilates is also a measure of  $\kappa$ .<sup>1,2,4,5</sup> Following the discovery<sup>6</sup> of the dependence of positron lifetimes in metals on the temperature  $T$ , Eqs. (1) and (2) and the equivalent relations for the concomitant changes in the angular correlation have been applied successfully to determine the energies of vacancy formation,<sup>7</sup>  $E_v$ . This is possible because  $\kappa \propto C = C_\infty \exp(-E_v/k_B T)$ . By Eq. (1), an independent determination of  $D_+$  would give, in addition, access to absolute vacancy concentrations by the positron method.

This Letter reports the first measurements of

$D_+$  in Fe, Co, Ni, W, and two FeNi alloys in the form of fine-grained powders. As in the earlier investigation of positron diffusion in small insulators,<sup>8</sup> the idea underlying the metal experiments is as follows. In powder grains small compared to the positron range, positrons emitted from a <sup>22</sup>Na source slow down with uniform probability throughout the grains to thermal energies in times  $\ll \tau_c$ . The positrons diffuse until they either (i) annihilate in the metal bulk with lifetime  $\tau_c$ , or (ii) are trapped at a rate  $\kappa$  in defects and annihilate there with lifetime  $\tau_d > \tau_c$ , or (iii) reach the surface where they are trapped into surface states and annihilate with lifetimes  $\tau_s > \tau_d$ . Exact formulas for the intensity  $\Phi$  of the lifetime component due to positrons reaching the surface have been derived in Ref. 2. They can be approximated within the experimentally attainable accuracy by the analytical form

$$\Phi = 1 - \exp\{-1.1 Sd[f(\bar{\tau})D_+\tau_c]^{1/2}/g_s\}, \quad (3)$$

where, with Eq. (2),

$$f(\bar{\tau}) = [1 + (\bar{\tau} - \tau_c)/g_s^2(\tau_d - \bar{\tau})]^{-1}. \quad (4)$$

$S$  denotes the specific powder surface in square centimeters per gram,  $d$  denotes the specific gravity of the metal in grams per cubic centimeter, and  $g_s = (1 - \tau_c/\tau_s)^{1/2}$ .

Positron lifetime spectra were measured at room temperature for the seven metal powders listed in Table I.<sup>9</sup> The coincidence apparatus used earlier was improved by incorporating a

TABLE I. Positron lifetime characteristics in metal powders. The lifetimes are accurate to  $\pm 5\%$ , the intensities to  $\pm 10\%$ .

Material	$r_s$ (a.u.)	$S^c$ ( $10^4$ cm <sup>2</sup> /g)	$Sd$ ( $10^6$ cm <sup>-1</sup> )	$\tau_1$ (nsec)	$\tau_2$ (nsec)	$I_2 = 1 - I_1^f$
Fe <sup>a</sup>	2.1	4.0	0.32	0.21	0.45	0.23
Fe <sup>b</sup>	2.1	0.48	0.04	0.19	0.45	0.04
Co <sup>c</sup>	2.1	4.35	0.39	0.20	0.37	0.29
Ni <sup>a</sup>	2.1	3.45	0.31	0.16	0.31	0.48
W <sup>d</sup>	1.6	0.70	0.135	0.21	0.45	0.09
Fe <sub>0.9</sub> Ni <sub>0.1</sub> <sup>a</sup>	2.1	12.8	1.02	0.21	0.40	0.42
Fe <sub>0.45</sub> Ni <sub>0.55</sub> <sup>a</sup>	2.1	7.3	0.61	0.20	0.35	0.38

<sup>a</sup> Powders kindly supplied by Dr. P. Dugleux, Ecole des Mines, Paris; the method of preparation is described in J. L. Doremieux, P. Dugleux, and G. Ciseron, *Silicates Ind.* **37**, 125 (1972).

<sup>b</sup> Manufactured by Office National Industriel de l'Azote, Toulouse, France.

<sup>c</sup> Manufactured by Ugine-Carbone, Grenoble, France.

<sup>d</sup> Purchased from Koch-Light, Colnbrook, Bucks, England.

<sup>e</sup> Ref. 10.  $S$  values are reproducible to  $\pm 2\%$ .

<sup>f</sup> All but the Fe ( $Sd = 0.04$ ) and W powders show third components of relative intensities  $I_3 = (2-8) \times 10^{-3}$  and lifetimes  $\tau_3 \approx 13$  nsec.

time-to-pulse-height converter of a design to be described elsewhere. The spectra were resolved into two components, with the intensities and lifetimes given in Table I. We identify  $\tau_1$  with the mean lifetime in the solid  $\bar{\tau}$ ,  $I_2$  with  $\Phi$ , and  $\tau_2$  with the lifetime in surface states,  $\tau_s$ . In independent measurements on well-annealed Fe slabs we found the positron lifetime to be 0.16 nsec, which we identify with  $\tau_c$ . We chose  $\tau_d = 0.22$  nsec as a suitable upper limit of  $\bar{\tau}$ , based on our measurements on all powders. By rearranging Eq. (3), one can deduce  $D_+$  from the slope of a plot of the quantity  $Y \equiv [g_s/1.1f(\bar{\tau})^{1/2}] \times \ln[(1 - I_2)^{-1}]$ , composed only of experimental positron lifetime data, versus the independently determined<sup>10</sup> powder variable  $Sd$ . As Fig. 1 shows, the locus of the points is a straight line, with the slope  $(D_+\tau_c)^{1/2} = 1.25 \times 10^{-6}$  cm, corresponding to a positron diffusion constant  $D_+ = (1.0 \pm 0.5) \times 10^{-2}$  cm<sup>2</sup>/sec at temperature  $T = 300^\circ\text{K}$ . This is the principal experimental result of our investigation. It offers the possibility of absolute vacancy-concentration measurements in metals with positrons.

The identification of  $I_2$  with  $\Phi$  as given by Eq. (3) assumes that the surface acts as a totally absorbing positron sink. If so, taking the internal-reflection coefficient  $r$  of the surface to be zero, one obtains from the data a lower limit for the bulk diffusion constant  $D_+$ . If  $r > 0$ , we have approximately  $I_2 = (1 - r)\Phi/(1 - r\Phi)$ . The observation  $I_2 \sim 0.3$  (Table I) implies that  $0 \leq r < 1$  and  $I_2 \leq \Phi < 1$ . The influence of  $r$  on the value of  $D_+$

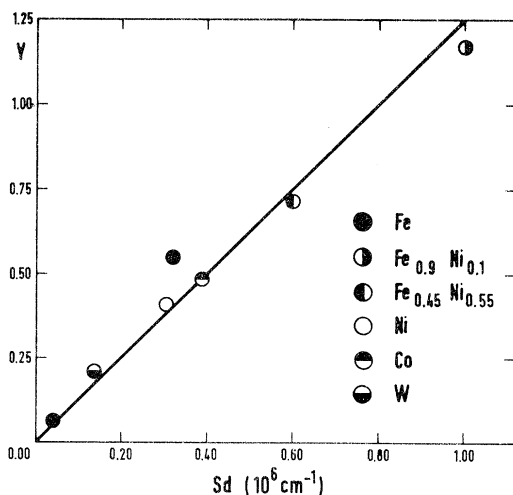


FIG. 1. The function  $Y$  discussed in the text versus  $Sd$ . The slope of the best straight line through the experimental points has the value  $(D_+\tau_c)^{1/2} = 1.25 \times 10^{-6}$  cm.

extracted from  $I_2$  data can be expressed as

$$\frac{D_+(r)}{D_+(r=0)} = \left\{ 1 + \frac{\ln[1 + rI_2/(1 - r)]}{\ln[(1 - I_2)^{-1}]} \right\}^2.$$

If  $r = 0.5$ , the value of  $D_+(r)$  is larger by a factor of 3 than the value of  $D_+(r=0)$  given above; if  $r = 0.8$ ,  $D_+(r)$  is larger by a factor of 10. We conclude that compared to the compounded uncertainties introduced by the lifetime data,  $D_+$  as deduced here is insensitive to the effects of internal reflections at the grain surfaces as long as the reflection coefficient is less than 0.5.

If we set  $D_+ = \frac{1}{2}\lambda v_{\text{th}}$ , where  $\lambda$  is a mean free scattering length and  $v_{\text{th}} = (2k_B T/m_+)^{1/2}$ , our  $D_+$  values imply that  $\lambda \sim 0.5 \text{ \AA}$  is smaller than the interatomic distance in a metal. We can gain some first theoretical understanding of  $D_+$  by estimating the effect of electron density fluctuations on the positron in terms of the uniform jellium model of a metal with interelectronic distance  $r_s$  and Fermi momentum  $k_F = (9\pi/4)^{1/3}r_s^{-1}$ . Here and in the following, all quantities are given in atomic units except where stated otherwise. Then,  $\lambda = v_{\text{th}}\tau_{\text{fl}}$ , where the time characteristic of electron-density fluctuations,  $\tau_{\text{fl}}$ , varies with the cutoff wave number  $k_c$  and the Fermi velocity  $v_F$  in the electron gas as  $\tau_{\text{fl}} \approx 2\pi/k_c v_F$ . Setting  $k_c = k_F$  for definiteness, we obtain  $\lambda \approx 0.1 \text{ \AA}$  in approximate agreement with experiment. More specifically, let the metal contain a positron as a plane wave with wave vector  $\vec{k}_+$  such that  $\langle k_+^2 \rangle = 2m_+k_B T$ . The effective positron mass  $m_+$  is of the order of  $1.25m_0$ .<sup>11</sup> We approximate the incoherent elastic scattering of the positron with electrons and phonons in terms of a local, constant scattering time  $\tau_{\text{sc}}$ . In the limit of small momentum transfer, this plane-wave model<sup>12</sup> yields  $D_+ = \frac{1}{2}\tau_{\text{sc}}\langle v_+^2 \rangle = \tau_{\text{sc}}k_B T/m_+$ , where  $\langle v_+^2 \rangle$  is the quantum-mechanical average over the positron velocity-velocity correlation tensor. The scattering time  $\tau_{\text{sc}} = (\tau_{+e}^{-1} + \tau_{+ph}^{-1})^{-1}$  depends on positron-electron scattering, with scattering time  $\tau_{+e}$ , and on positron-phonon scattering, with  $\tau_{+ph}$ .

The conduction-electron gas screens the positron charge over a distance  $\sim r_s^{1/2}$ . In the interaction with a positron, the wave function of an electron of wave number  $k$ , and momentum and spin states  $\mu = (l, m, s)$  relative to the scattering center, incurs the phase shift  $\eta_\mu(k)$ . The screening electron charge builds up because electrons in the state  $(k, \mu)$  are retained by the positron for a time  $\tau_\mu(k) = (2/v) d\eta_\mu/dk$  ( $v = \text{velocity}$ ),<sup>13</sup> corresponding to an electron scattering rate  $\tau_\mu^{-1}(k)$

on the positron "impurity." The resulting charge increments per unit interval of  $k$  are  $n_\mu = \pi^{-1} d\eta_\mu / dk$ ; they obey the Friedel sum rule  $\sum_\mu \int_0^{k_F} n_\mu dk = 1$ .<sup>14</sup> The weighted positron-electron scattering rate for the one electron that interacts on the average with the positron at any given time becomes

$$\tau_{+e}^{-1} = \int_0^{k_F} n_\mu(k) \tau_\mu^{-1}(k) dk = v_F k_F / 4\pi. \quad (5)$$

In this approximation,  $\tau_{+e}$  is a constant independent of  $\mu$ . With  $v_F = k_F \approx 2r_s^{-1}$ ,

$$\tau_{+e} = \pi r_s^2 \text{ a.u.}, \quad (6)$$

where 1 a.u. =  $2.42 \times 10^{-17}$  sec. More detailed considerations of the momentum dissipation rate by the partial-wave analysis under the phase-space restrictions of an electron gas are required for an *ab initio* calculation of  $\tau_{+e}$ .

The well-known formula<sup>15</sup>

$$\tau_{+ph} = 2^{3/2} \pi^{1/2} u_l^2 d / 3m_+^{5/2} E_d^2 (k_B T)^{3/2} \text{ a.u.} \quad (7)$$

serves for an orientation as to positron-phonon scattering times;  $u_l$  is the velocity of long-wavelength compressional waves in the solid. An upper limit of the deformation-potential constant  $E_d$  is the positron binding energy in a vacancy,  $\sim 1$  eV.<sup>16</sup> The values of  $\tau_{+ph}$  typical for metals exceed  $\tau_{+e}$ , so that under normal conditions positron-phonon scattering should not be the diffusion-limiting process.

With Eq. (6), then, one expects that in metals (m)

$$D_+^m(T) \approx (\tau_{+e} / m_+) k_B T \approx (3.25 \times 10^{-3}) r_s^2 (T / 300^\circ \text{K}) \text{ cm}^2 / \text{sec}. \quad (8)$$

In the powders considered here,  $r_s = 2.1$ , and Eq. (8) yields  $D_+^m(300^\circ \text{K}) = 1.4 \times 10^{-2}$  cm<sup>2</sup>/sec. While the quantitative agreement with experiment is fortuitous, the order of magnitude appears to be correct.

In solids with an energy gap  $E_g \gg k_B T$ , the positron-electron scattering rate may be reduced drastically, as a result of the reduction in conduction-electron density, by a factor of  $\sim \exp(-E_g / 2k_B T)$ . Positron-phonon scattering would then dominate. In this limit, the diffusion constant in insulators and semiconductors (s),

$$D_+^s(T) \approx (\tau_{+ph} / m_+) k_B T \approx (1 \times 10^2) (300^\circ \text{K} / T)^{1/2} \text{ cm}^2 / \text{sec}, \quad (9)$$

is larger than  $D_+^m$  by a factor of  $\sim 10^4$ . Recent

observations on Ge powders,<sup>17</sup> when analyzed along the lines presented here, could well be consistent with this conclusion.

We attribute the component  $I_2$  to positrons that annihilate in surface states with lifetimes  $\tau_s \approx \tau_2$ . Let the electron density  $\rho(x)$  near the surface (located at  $x=0$ ) of a metal, with bulk density  $\rho_0$  at  $x \ll 0$ , be approximated by<sup>18</sup>

$$\rho(x) = 3/4\pi r_s^3(x) = \rho_0 / 2 \{1 - \text{sgn}(x) [1 - \exp(-|x|/a_s)]\}, \quad (10)$$

where  $a_s \sim r_s^{1/2}$  is the surface screening length. The lifetime of a positron located at  $x$  can be estimated from the relation<sup>19</sup>

$$\tau_s(x) \approx \frac{1}{12} r_s^3(x) \{1 + f(E_g) [r_s^3(x) + 10] / 6\}^{-1} \text{ nsec.} \quad (11)$$

For the "clean" surfaces of Ni,  $f=1$ , one finds that  $\tau_s = \tau_2$  when  $x \approx +a_s$ . Recently, Smith, Ying, and Kohn<sup>20</sup> reported a Hartree-Fock calculation of the interaction of a proton with a metal surface which has an energy minimum of  $\sim 1$  eV just at  $\sim +a_s$ .<sup>21</sup> The larger values of  $\tau_2$  in Fe and Fe alloys presumably result from the fact that their surfaces are passivated by the formation of oxides. This would reduce the enhancement ( $f < 1$ ),<sup>19</sup> and hence increase  $\tau_s$ , even if the positrons are trapped at the same surface position.

\*Work supported by the French Atomic Energy Commission and the U.S. National Science Foundation.

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<sup>9</sup>The mean depth of positron penetration,  $\alpha^{-1}$ , is  $\alpha^{-1} \approx [40d(\text{g/cm}^3)]^{-1}$  cm. The average radius  $R$  of the powder grains is  $R \approx \mu(Sd)^{-1}$  cm, where  $\mu = 2$  or 3 for cylindrical or spherical grains, respectively. Since  $\alpha R \sim 10^{-2} \ll 1$  in all powders investigated, the probability of positron deposition is constant throughout each grain.

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<sup>16</sup>Use of Eq. (7) to explain  $D_+$  in metals (Ref. 7) requires unreasonable values of  $E_d$  ( $> 50$  eV); the result-

ing dependence  $D_+^m \propto T^{-1/2}$  remains unconfirmed by experiment; cf. D. E. O'Connors and J. C. Bowler, Phys. Lett. 43A, 395 (1973).

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## Surface Forces and the Jellium Model

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(Received 13 July 1973)

Analytic results are derived for the surface properties of jellium. The surface potential relative to its bulk value is determined exactly in terms of the bulk properties of the uniform electron gas. This result is independent of any assumption concerning the electron energy density functional in an inhomogeneous system and depends only on the energy density in the homogeneous solid. We discuss the basic inapplicability of dispersion-type calculations of the surface energy.

While progress has recently been made in understanding the surface properties of solids, no exact results have been obtained in this area. Elaborate numerical calculations of the electronic ground state in the fixed background potential of the ions have been presented.<sup>1,2</sup>

In this Letter we present the first exact result relevant to the surface properties of the jellium model. We calculate the surface potential of the jellium relative to its bulk value exactly. No use is made of an approximate form for the energy density functional of the inhomogeneous electron system. Our results depend only on the bulk properties of the jellium, and we obtain explicit results for an arbitrary form of the bulk energy density.

We now consider the jellium model, with the jellium background split into two parts separated by a distance  $2D$ , as in Fig. 1. The force acting on an infinitesimal slab of background charge is simply  $q\bar{E}$ , where  $q$  is the total charge of the slab and  $\bar{E}$  is the expectation value of the electric field at the slab position.<sup>3</sup> This is the well-known result of the Feynman-Hellman theorem.<sup>4</sup> Since

the electric field is generally nonzero, the jellium cannot conceivably be in equilibrium. If in fact we had considered a fixed discrete ionic background, the ion positions would be determined by just this condition, namely that they be at potential energy minima.

The force that one half of the split jellium ex-

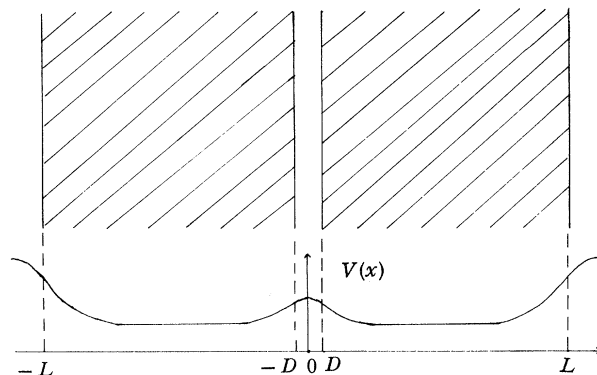


FIG. 1. Schematic representation of two parts of a solid separated by a gap  $2D$ . Lower curve represents electron electrostatic potential energy.