Temperature dependence of positron diffusion in cubic metals

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We have applied a slow-positron-beam technique to extract quantitative results for the positron diffusion coefficient in high-purity defect-free Mo, Al, Cu, and Ag single-crystal samples in the temperature range 20–1400 K. The value of the diffusion coefficient D_+ at 300 K is 1–2 cm^2/s, and it follows a power law $D_+ \propto T^{-\alpha}$ as a function of the temperature T with $\alpha = 0.5$–0.6 in the studied temperature range. The results show that positron diffusion is largely limited by scattering from longitudinal acoustic phonons. A good agreement with band-structure calculations is obtained with the positron effective mass $m^* \approx 1.5$. The terms affecting the temperature dependence of the sum of the positron and electron chemical potentials are extracted.

I. INTRODUCTION

Precise knowledge of the positron diffusion coefficient D_+ in solids is of great interest for fundamental understanding of positron interactions in solids. The absolute value and the temperature dependence of D_+ are measures of the nature and strength of the various positron-solid interaction mechanisms.

The use of the diffusion picture supplies an experimental test for the validity of the Nernst-Einstein relation for an interacting positron through a comparison to theory. Quantitative information on D_+ as a function of temperature is also needed for applied techniques such as defect profiling with monoenergetic positrons, or positron studies of layered structures (for a review, see Ref. 1).

In positron-beam experiments keV-range positrons are implanted into the sample. After thermalization a fraction of them diffuses back to the surface. In defect-free metals positrons can only annihilate in the bulk or at the surface region. The difference between these two states provides the information necessary to obtain D_+. For the proper interpretation of the positron experiments at surfaces it is also essential to know positron diffusion properties in the bulk.

Positron diffusion is expected to be limited by acoustic phonon scattering,[2] which leads to the temperature dependence for the diffusion coefficient $D_+ \propto T^{-1/2}$. Band-structure calculations for most metals yield approximately $D_+ \approx 1$ cm^2/s at room temperature.[3,4]

Among the first experimental studies on positron diffusion were positron mobility experiments performed on Ge and Si.[5,6] In metal and semiconductor powders very small values of D_+ were found (for a review, see Ref. 7). Early experimental positron-beam results from metals did not support the simple theory of positron diffusion governed by scattering from acoustic phonons. The temperature dependence of D_+ was found to be significantly stronger than $T^{-1/2}$ (Ref. 8). Below 160 K in Al, values of D_+ were reported that are substantially lower than expected from the high-temperature data.[8] This was attributed to positron localization into weakly

bound states at low temperatures. However, at temperatures above 300 K Huomo et al.[9] showed that in Mo(111) the deviation from the theory can be explained in terms of incorrect treatment of epithermal positron effects and the implantation profile parametrization.

In this report we publish slow positron beam results on positron diffusion for a temperature range 20–1400 K and demonstrate that the agreement between our experiments and theory holds for several cubic metals down to 20 K. No significant deviations from the power law $D_+ \propto T^{-\alpha}$ are observed even at the lowest temperatures, with $\alpha = 0.5$–0.6. Several factors affecting the precise value of the power α are discussed. Furthermore, values for the deformation potential parameter E_d are derived from our results. A comparison with other results is done in order to estimate the positron effective mass and the effect of lattice vibrations on the electrostatic potential of positrons in metals.

In the next section we shortly review the theoretical treatment adopted by Bergersen et al.[2] to describe the interaction of positrons with longitudinal acoustic phonons. The experimental technique is presented in Sec. III, and a description of our method of data analysis in Sec. IV. The results for the temperature dependence and the absolute values of the positron diffusion coefficients in several cubic metals are shown in Sec. V. In Sec. VI we discuss the implications of the results to the theory of positron motion and positron potential in metals. Finally, Sec. VII concludes the paper.

II. THEORETICAL ASPECTS

Thermalized positron motion in solids is usually treated with the diffusion theory, where it is required that the positron mean free path be much shorter than the region under inspection. In positron-beam experiments with keV incident positron energies this requirement is satisfied. Furthermore, it is assumed that the thermal positron distribution follows Maxwell-Boltzmann statistics and that the scattering events are almost entirely elastic and isotropic. In high-purity defect-free metals also these assumptions seem reasonable.[10,11]

In the Nernst-Einstein relation, the positron diffusion coefficient D_+ is written in terms of the Boltzmann constant k_B, temperature T, the electron charge e, the positron mobility μ, the average scattering rate $\langle \dot{n} \rangle$ and the positron effective mass m^*:

$$D_+ = \frac{k_B T \mu}{e} = \frac{k_B T}{m^* \langle \dot{n} \rangle} \ . \tag{1}$$

In a metal, a thermalized positron can scatter inelastically from conduction electrons, elastically from impurities, and quasielastically from phonons. Scattering from conduction electrons would lead to a temperature dependence $D_+ \propto T^{-1}$ and impurity scattering to $D_+ \propto T^{+1/2}$ (Ref. 2). However, for several simple metals Bergersen *et al.*[2] estimated phonon scattering to dominate the scattering rate over electron scattering. Furthermore, only in the limit of high impurity concentration and low temperatures does impurity scattering become competitive with phonon scattering. Consequently, the dominant contribution comes from the longitudinal acoustic phonons, which Bergersen *et al.* treated by using a deformation potential.[12] In the high-temperature limit $(T > 10$ K) an expression for the scattering rate is obtained[13] (see also Sec. VI B):

$$\langle \dot{n}_{ph} \rangle = \frac{\sqrt{2} \pi^{3/2}}{4} \frac{E_d^2 m^{*3/2} (k_B T)^{3/2}}{\hbar^4 \langle c_{ii} \rangle} \ , \tag{2}$$

where $\langle c_{ii} \rangle$ is the elastic stiffness constant associated with longitudinal waves averaged over all the directions of propagation and E_d the deformation-potential parameter. According to Eqs. (1) and (2), the positron diffusion coefficient varies with temperature as $D_+ \propto T^{-1/2}$.

E_d is defined as $V(\partial E / \partial V)$ where $E(V)$ is the total energy of the crystal in volume V, with a positron in its lowest Bloch state. It follows that E_d can be expressed as

$$E_d = V \frac{d\mu_+}{dV} + V \frac{d\mu_-}{dV} \ . \tag{3}$$

The first term describes the volume dependence of the positron chemical potential which includes the positron zero-point energy E_0 and the electron-positron correlation energy E_{corr}. Since the electron chemical potential μ_- must remain constant throughout the crystal there will be electrostatic potentials compensating for the induced charge transfer. This is the origin of the second term in Eq. (3).

III. EXPERIMENTAL TECHNIQUE

The measurements were performed with a magnetically guided monoenergetic positron beam, with the beam intensity up to 5×10^6 e^+/s.[14] The positron diffusion coefficient was evaluated from positron back-diffusion measurements by observing changes in the Doppler broadening of the annihilation line or the fraction of positronium (Ps) emitted from the surface, as a function of incident positron energy (0.13–25 keV). The line-shape parameter S and the Ps yield were measured with standard methods[1] by observing annihilation γ-ray spectra with a

high-purity Ge detector, with a positively biased grid in front of the sample to turn the reemitted positrons back to the surface.

The samples were electrolytically polished single crystals. The (99.9999%+)- purity Al(110) sample was delivered by Cominco Co, (99.9999%+)- purity Cu(111) by Tampere University of Technology, (99.9999%+)- purity Ag(111) by Metal Oxides and Crystals Co., and (99.99%+)- purity Mo(110) and Mo(111) samples by Materials Research Co. For the annihilation line-shape parameter measurements, the surfaces were not cleaned *in situ* in order to avoid Ps formation[15] and the pressure in the sample chamber was typically 1×10^{-8} mbar. For the Ps fraction measurements, the surfaces were cleaned *in situ* with standard Ar^+ sputtering and annealing cycles and possible contamination was monitored with a retarding field Auger electron analyzer. In the latter case, the pressure was typically 2×10^{-10} mbar and the surface condition did not change during a measurement at each temperature. At room temperature the surface contaminant concentration was observed to be less than 1 at. %.

When measuring the Ps yield (f_{Ps}) from a clean surface, at each incident positron energy typically 250 000 counts were collected to the 511 ± 7 keV annihilation peak, whereas during the line-shape parameter measurements, 1×10^6 events were recorded. At each temperature around 50 $S(E)$ or $f_{Ps}(E)$ points were collected. It took about 1 h and 2–9 hr to measure the Ps yield and the S parameter versus incident energy at each temperature, respectively.

Temperature was maintained with a closed-loop He cryocooler and an electron-beam heater and measured with a type-K (NiCr/NiAl) thermocouple attached to the sample surface. The thermocouple was calibrated against a Si diode and a Au/NiCr thermocouple, and the error below 30 K is estimated to be ± 5 K and at room temperature less than ± 2 K. Temperatures from 20 to 1400 K were measured in random order.

IV. DATA ANALYSIS

Thermal positron motion in solids can be described with the diffusion equation.[1] In monoenergetic positron-beam experiments the one-dimensional diffusion-annihilation equation yields the positron back-diffusion probability $J(E)$ at the surface for an incident positron energy E,

$$J(E) \propto \int_0^\infty P(z,E) e^{-z/L_+} dz \ , \tag{4}$$

where the z axis is taken perpendicular to the surface, $P(z,E)$ is the implantation profile and L_+ the positron diffusion length. The implantation profile can be expressed as $P(z,E) = (-d/dz)\{\exp[-(z/z_0)^m]\}$ with $z_0 = A_0 (E/\text{keV})^n$. We have chosen parameter values $m \approx 2$ and $n \approx 1.6$ based on Monte Carlo simulations[16] and direct measurement of the implantation profile shape and the incident energy dependence of positron implantation.[17] Electron-stopping experiments[18] indicate that parameter n might have an incident-energy dependence. We do not expect that a weak energy dependence of the

parameter n could affect our data analysis because back-diffusion measurements give emphasis to a rather narrow incident-energy regime. Parameter A_0 is a material parameter for which we have chosen $A_0 = (4.5 \ \mu g/ cm^2)/\rho$,[15,17] where ρ is the density of the material.

The diffusion length is related to the diffusion coefficient by the relation

$$L_+ = \sqrt{D_+ \tau_{\text{eff}}} \, , \quad (5)$$

where the effective lifetime τ_{eff} of a freely diffusing positron in a defect-free metal equals its bulk lifetime τ_b. At the temperature range used, the positron bulk lifetimes are assumed constant, and in Mo, Al, Cu, and Ag their values are 110 ps,[19] 163 ps,[20] 110 ps,[21] and 130 ps,[21] respectively.

The slow-positron beam technique is based on the assumption that both the Ps yield f_{Ps} and the annihilation line-shape parameter S are linear to the back-diffusion probability $J(E)$ of thermal positrons:[1]

$$f_{Ps}(E) = f_0 J(E) \quad (6)$$

and

$$S(E) = (S_{\text{surf}} - S_{\text{bulk}}) J(E) + S_{\text{bulk}} \, . \quad (7)$$

The parameter f_0 is the fraction of positronium formed from thermal positrons at the surface. The parameter S_{bulk} is a characteristic value of the bulk annihilation line shape at high incident positron energies and S_{surf} the corresponding value for annihilations at the surface region. The measured data are fitted with a method of least squares to Eqs. (6) and (7). The analyzing programs are described in Ref. 15.

Figure 1 shows measured S parameter values obtained from a Mo(110) sample as a function of incident positron
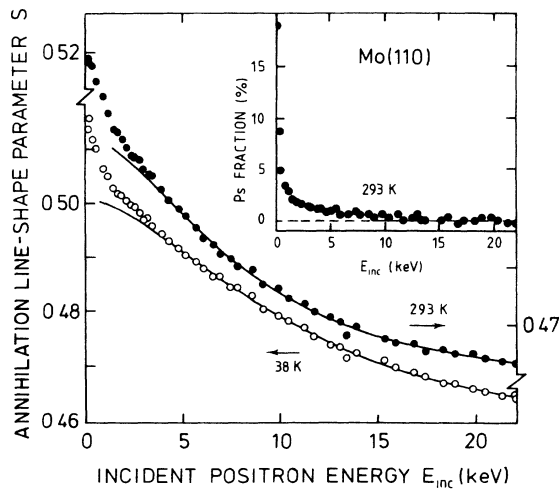

FIG. 1. Typical data for the measured annihilation line-shape parameter S vs incident energy at 38 and 293 K. The sample is a well-annealed Mo(110) crystal with a thin oxide overlayer. Solid lines are linear fits to Eq. (7) for the energy range 4–23 keV. In the inset a simultaneously measured Ps yield at 293 K is also shown.

energy at two temperatures, together with fits to Eq. (7). Low incident positron energies were omitted from the fit because (i) a significant fraction of the positrons returning to the surface have not thermalized[9,22,23,24] and (ii) the positron penetration depth is of the order of the mean free path and therefore the implantation profile is not accurately known and the diffusion equation is no longer valid.[15]

Equation (7) strictly holds only if no positronium is formed.[15] Therefore, in the annihilation line-shape measurement the Ps fraction has to be close to zero. At an uncleaned surface for higher energies than a few keV this condition is shown to be satisfied even at high temperatures (see inset of Fig. 1).

The lowest incident positron energy included in the fit was found by varying the minimum energy E_{min} until fits to Eqs. (6) and (7) gave stationary values for the parameters m, n, D_+, and the variance χ^2 (Refs. 9 and 15). Such an optimization of E_{min} was done for each sample at each temperature. The data from all temperatures have been analyzed with the same E_{min} for each sample, the used E_{min} being the largest value found for each material.

It would be logical to expect the minimum energy to increase with the density of the material, as in dense materials the positron needs more energy to penetrate to a certain depth. In the line-shape measurements this indeed is the case; the lowest energies E_{min} were 2, 4, and 6 keV for Al(110), Mo(110), and Cu(111), respectively. The corresponding threshold energies E_{min} in the Ps-yield measurements were 3.5, 4, and 3 keV for Al(110), Mo(110), and Ag(110). Now the situation is more complicated, as E_{min} for Ag is lower than for Al. This may be due to the effect of the positron work function ϕ_+ on the epithermal Ps yield. In Ag(111) ϕ_+ is positive whereas in Al(110) it is negative. Ps formation seems to be very sensitive to the energy levels at the surface.[25,26]

The implantation profile parameters were observed to be consistent with the earlier values $m \approx 2$ and $n \approx 1.6$. It was found that in Al and Mo the minimum of the variance in (m, n) space is located around $m = 2.0$ and $n = 1.55$ (see Ref. 15). These values were chosen to parametrize the implantation profile in the subsequent analyses.

V. RESULTS

The experimental results for the positron diffusion coefficient D_+ as a function of temperature in Mo, Al, Cu, and Ag are shown in Fig. 2. The extracted values of D_+ at 300 K are in all cases 1–2 cm²/s. It can also be seen that in all examined metals, the diffusion coefficient D_+ apparently follows a simple power law from about 20 K up to the threshold of thermal vacancy formation, where the lifetime of a positron in a delocalized state τ_{eff} in Eq. (5) becomes shorter.[1]

Two sets of line-shape measurements were run on Al(110). One set (filled squares in Fig. 2) gives significantly more reliable and consistent results for $D_+(T)$ than the other (filled circles). This is partly due to much higher beam intensity and count rate at the detector during the latter measurement which, in turn,
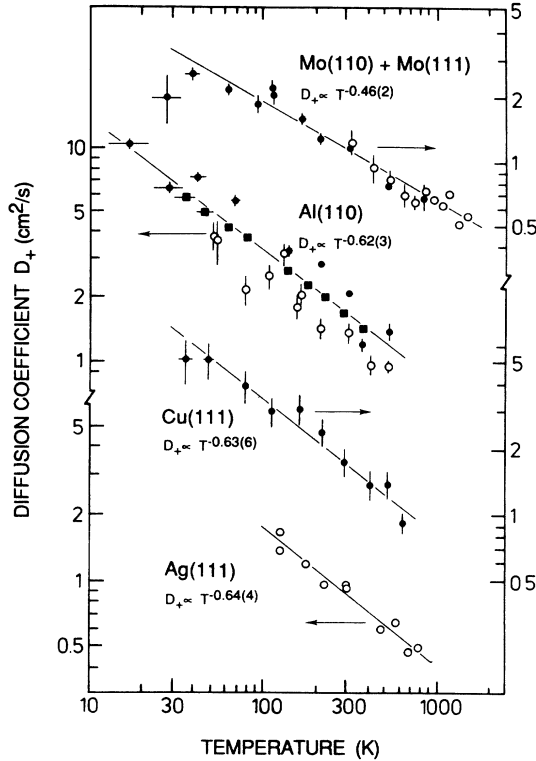
FIG. 2. The temperature dependence of the positron diffusion coefficient in Mo(110), Mo(111), Al(110), Cu(111), and Ag(111). Open symbols indicate Ps fraction measurements and solid ones annihilation line-shape measurements. The lines are fits to a power law $D_+ = D_0(T/300 \text{ K})^{-\alpha}$ for all the data points in the figure. The shown errors of D_+ are due to statistical errors of the S parameter or Ps fraction. The error bars for Ag(111) are of the size of the symbols shown. In addition, the evaluation of emitted Ps fraction requires a number of calibration parameters whose estimated systematic error contribution is also included.

sets much higher requirements for the data acquisition system. The Ps yield results of Mo(111) above 300 K in Fig. 2 are directly extracted from Ref. 9. For Mo(110) at lower temperatures the line-shape technique was applied, while the 780-K point is from the Ps fraction data, due to the strong Ps formation and consequent nonlinearity of the S parameter at this temperature (see Refs. 15 and 27).

The results of the annihilation line-shape parameter measurements for Mo(110) at low temperatures were combined with the Ps fraction measurements for Mo(111) at higher temperatures.[9] Both results fit very well the same line, verifying the equivalence of the two methods and the expected isotropy of diffusion with respect to the crystal orientation in close-packed cubic materials.

The measured D_+ values were least-squares fitted to a power law $D_+ = D_0 (T/300 \text{ K})^{-\alpha}$ for the whole temperature range. The resulting fits are drawn in Fig. 2 as solid lines and the extracted D_0 and α values are listed in Table I. The combined results for Al and Mo have been obtained by fitting all the $D_+(T)$ points from different runs together. It is seen that in all cases the power α slightly deviates from the predicted value $\frac{1}{2}$. However, in Mo and Cu the deviation is only 2σ, in Ag about 3σ, and in Al 4σ, where σ is the standard deviation of the power α. Due to the very good accuracy of the first Doppler measurement in Al(110), we feel that in Al the power α seems to deviate significantly from $\alpha = \frac{1}{2}$ whereas in other metals possible deviations are less significant. In any case, values of α are around $\alpha \approx 0.6$ in all fcc metals studied.

VI. DISCUSSION

A. Comparison of our results with the deformation-potential theory

The results fit rather well to the predicted power law $D_+ \propto T^{-\alpha}$ with $\alpha \approx \frac{1}{2}$ from about 20 K to more than

TABLE I. The results of the fit of the positron diffusion coefficient D_+ to a power law $D_+ = D_0(T/300 \text{ K})^{-\alpha}$ for several cubic metals in the temperature range indicated. The diffusion length L_+ is defined in Eq. (5). The errors consist of contributions from statistical and systematic sources. The systematic errors are estimated to be in the worst case (second line-shape run of Al) about 60% of the total errors indicated. Dop = Doppler broadening measurement of the annihilation line-shape parameter S. Ps = Ps fraction measurement.

Material surface	Method	Power α	Temperature range (K)	D_0 (cm²/s)	L_+(300 K) (Å)
Al(110)	Dop-1	0.60(1)	34–360	1.63(9)	1630(50)
	Dop-2	0.58(6)	16–500	2.0(5)	1800(300)
	Ps	0.63(9)	50–505	1.3(6)	1500(300)
Mo(110)	Dop	0.46(5)	27–780	1.1(3)	1100(100)
Mo(111)	Ps	0.47(8)	300–1400	1.1(6)	1100(300)
Combined results					
Al(110)	Dop+Ps	0.62(3)	16–505	1.7(2)	1600(100)
Cu(111)	Dop	0.63(6)	35–620	1.7(5)	1400(200)
Ag(111)	Ps	0.64(4)	120–770	0.9(2)	1100(100)
Mo(110,111)	Dop+Ps	0.46(2)	27–1400	1.1(1)	1100(70)

TABLE II. Comparison of the extracted deformation-potential parameter E_d with theoretical and other experimental results. In first two columns E_d^{expt} are calculated by substituting our experimental results for D_+ from Table I at 300 K into Eqs. (1) and (2) with elastic constants $\langle c_{ii} \rangle \approx \frac{1}{2}(c_{11}+c_{12}+2c_{44})$ from Ref. 43, and using values of m^* as indicated. Theoretical estimates E_d^{theor} are based on the calculation of electron and positron energy levels at 300 K. Other experimental results E_d^{expt} are obtained from work-function measurements at various temperatures, with the lattice vibrational term in Eq. (8) assumed zero (for discussion, see text).

Metal	$\langle c_{ii} \rangle$ $(10^{11}$ N/m$^2)$	$E_d^{\text{expt}}(D_+)$/eV $m^*=1.5m_e$	$m^*=1.2m_e$	E_d^{theor} (eV)	$E_d^{\text{expt}}(d\phi_0/dT)$ (eV)
Al	1.12	-6.7	-8.8	-7.70,[a] -8.6,[b] -9.63[c]	$-11.7(5)$,[d] -13.4[e]
Cu	2.21	-9.4	-12	-9.45,[a] -10.42[c]	-14.6,[e] $-14.5(3)$[f]
Ag	1.53	-11	-14	-9.48[a]	
Mo	4.21	-16	-21	-14.3[a]	

[a]Boev et al. (Ref. 3)
[b]Bergersen et al. (Ref. 2).
[c]Farjam and Shore (Ref. 4).
[d]Gullikson and Mills (Ref. 35).
[e]Rosenberg et al. (Ref. 40).
[f]Gidley (Ref. 41).

1000 K. This is in agreement with the theory where thermal positron motion is limited by scattering from acoustic phonons, as described in Sec. II. The diffusion coefficient D_+ at 300 K, about 1 cm^2/s, is in accord with the deformation-potential theory, where the coupling of the positron with acoustic phonons is expected to be weak.[11]

A quantitative comparison with the theory can be done via the deformation-potential parameter E_d which can be readily calculated, substituting the extracted results for D_+ at 300 K into the expressions (1) and (2). In Eq. (1) a quantity m^* denoting the positron effective mass is needed. Estimates for m^* vary between $1.2m_e$ and $1.5m_e$.[28] The calculated E_d values with $m^*=1.5m_e$ and $m^*=1.2m_e$ are presented in Table II, together with some theoretical and independent experimental results. We can see that the use of $m^*=1.5m_e$ provides a good quantitative agreement for our results with the ones based on the deformation-potential theory and band-structure calculations.[3,4] Our experimental values of D_+ quantitatively reproduce the systematic trends for different materials predicted by the deformation-potential theory.

B. Potential sources for deviations from the $T^{-1/2}$ law

No deviation from the power law $D_+ \propto T^{-\alpha}$ is observed even at the lowest temperatures. This is a clear indication that in pure defect-free metals no shallow traps are observed at low temperatures, as suggested earlier.[8] The D_0 values in Table I are in general somewhat higher than results from the earlier positron beam experiments.[29–32] The discrepancy between the present and earlier results is due to the effect of epithermal positrons at the lowest incident positron energies, which have now been omitted from the data. Furthermore, particularly in dense materials, much higher incident positron energies (>20 keV) should be used in order that we can unambiguously identify the shape of the back-diffusion current and the values of the diffusion coefficient. Third reason for the discrepancy is the earlier use of an exponential

implantation profile ($m=1$) which has been shown to be incorrect.[17] These points are discussed in detail elsewhere.[15] The values obtained for the power α were found insensitive to small variations in the implantation depth parameter n ($|\partial\alpha/\partial n| \leq 0.2$).

Numerical calculations in the deformation-potential model using the full Bose-Einstein distribution for the phonon densities and the Maxwell-Boltzmann distribution for the positron energies show that the phonon absorption and emission processes give rise to equal contributions to D_+.[33] The power law $D_+ \propto T^{-1/2}$ holds down to about 10 K, below which the values of D_+ sharply rise. In Eq. (2), a high-temperature limit of these calculations is shown.[13] A factor $(1-\cos\theta)$, where θ stands for the scattering angle, has been omitted in calculating the positron phonon scattering rates. The inclusion of this factor leads to numerical corrections below about 20 K.[33] Thus, in the temperature regime studied, no deviations from the power law $D_+ \propto T^{-1/2}$ are expected due to the phonon distribution.

In the fcc crystals studied (Al, Cu, and Ag), the power α is slightly higher than the value predicted by the deformation-potential theory. This is rather surprising since, due to the implicit long-wavelength phonon approximation, the deformation-potential theory should be particularly suited for particles of low energy,[12,33] such as a thermalized positron. The inclusion of transverse phonon scattering in the theory does not explain why the temperature dependence for D_+ in fcc crystals is stronger than in bcc (Mo) crystals. The cross section for positron transverse phonon scattering depends on the probability of phonon umklapp processes (for electrons, see Ref. 34) and can be estimated to increase as the lengths of the shortest reciprocal-lattice vectors decrease.[33] This leads to a more pronounced effect for a less densely packed structure, such as a bcc crystal in comparison to a fcc crystal. Taking into account the actual lattice parameters, the power α could be expected to be slightly larger in Mo than in Al, Cu, and Ag.

The deformation-potential parameter E_d has recently

been estimated to be independent of temperature below 300 K.[35] This excludes the suggested possibility of a temperature-dependent E_d significantly affecting the D_+ power law.[8] Furthermore, band-structure calculations show that in Al E_d depends only weakly on temperature and at 0 K it is 2% larger than at 300 K.[36] This causes a change in α from $\alpha \approx 0.5$ to $\alpha \approx 0.49$.

Scattering from conduction electrons would lead to a dependence $D_+ \propto T^{-1}$ for the positron diffusion coefficient. Nonadiabatic screening effects result in a $T^{-0.8}$ dependence for a positive muon diffusion in solids.[37] For positron diffusion this effect is estimated to be very weak.[38] However, now that more accurate experimental values of α are available, it might be worthwhile to reconsider the influence of positron-electron interactions to the temperature dependence of positron diffusion.

Above 100 K, the elastic stiffness constants $\langle c_{ii} \rangle$ in Eq. (2) decrease linearly with temperature.[39] Using the $\langle c_{ii} \rangle$ data between 30 and 300 K we obtain, in this temperature range, the power $\alpha \approx 0.51$ in Mo and $\alpha \approx 0.52$–0.53 in the other metals under study. Thus we see that the temperature dependence of the stiffness constants explains a part of the observed deviation from $D_+ \propto T^{-1/2}$.

Yet another factor affecting the power law of D_+ arises from the possible effect of temperature on the positron effective mass m^*. From Eqs. (1) and (2) it can be seen that a change from $m^* \approx 1.4$ at 30 K to $m^* \approx 1.5$ at 300 K would explain the high value of the power $\alpha \approx 0.6$ instead of the predicted $\alpha \approx 0.5$. Unfortunately it is very difficult to estimate $m^*(T)$ either numerically or experimentally.

From Fig. 2 one can see that a slope of $\alpha \approx 0.6$ could fit to the D_+ results from Mo for $T > 100$ K. At low temperatures the D_+ values would then be lower than expected from high temperatures. The effect could be ascribed to impurity scattering, which might affect particularly the results in our Mo sample, since there the impurity concentration is the highest among the samples under study (substitutional W 70 ppm, interstitial C 10 ppm). However, applying the formula in Ref. 2 for the positron scattering rate $\dot{n}_i$ from impurities and estimating for the host-impurity potential difference 1 eV, we get $\dot{n}_i / \dot{n}_{ph} < 0.03$ at all temperatures above 30 K. Thus we can conclude that unless the near-surface impurity concentration is much higher than in the bulk, impurity scattering does not affect our results significantly.

C. Comparison of our results with work-function measurements

When comparing our results with other experiments (Table II), we note that both values of m^* yield lower values for E_d than the direct work-function measurements.[35,40,41] The reason for the discrepancy lies in the definition of the quantities. At constant pressure, the temperature dependence of the sum of the electron and positron work functions ϕ_0 can be expressed as[42]

$$\frac{d\phi_0}{dT} = \gamma V \left[\frac{\partial \phi_0}{\partial V} \right]_T + \left[\frac{\partial \phi_0}{\partial T} \right]_V , \tag{8}$$

where γ is the thermal volume expansion coefficient. From the definition of the deformation-potential parameter it follows that the first term is $-\gamma E_d$. The second term is due to the effect of lattice vibrations on the internal electrostatic potential.[3]

Boev *et al.*[3] have shown that there are no *a priori* reasons to expect that this term is significantly smaller than the first one. However, Gullikson and Mills[35] assumed this term to be vanishingly small, and based on their measurements of ϕ_0 with temperature they evaluated E_d directly from the first term in Eq. (8). The tabulated E_d values from Refs. 39 and 40 have been extracted similarly. The experiment of Gullikson and Mills[35] shows that $d\phi_0/dT$ is proportional to the thermal expansion of the lattice. As they point out, this indicates that the lattice vibrational term either vanishes or it has the same dependence on temperature as γ. For Al, our new result combined with the experiment of Gullikson and Mills[35] gives for the second term of Eq. (8) 0.2–0.4 meV/K, depending on the choice of the positron effective mass. This is by no means negligible with the first term which, by applying our result for E_d, can be estimated to vary between 0.4 and 0.6 meV/K. The comparison of the results with those of Rosenberg *et al.*[40] gives similar large values for the temperature gradient of the lattice vibrational term in Cu, 0.1–0.3 meV/K. The term $-\gamma E_d$ in Cu is 0.4–0.6 meV/K, respectively. It would be interesting to test our experimental results for $(\partial \phi_0 / \partial T)_V$ against theoretical calculations.

VII. SUMMARY

Temperature-dependent positron-diffusion coefficients have been measured for several cubic metal single crystals. Our results show that positron diffusion in Al, Cu, Ag, and Mo is mainly limited by scattering from longitudinal acoustic phonons. The power law $D_+ \propto T^{-\alpha}$ holds from 20 K to the limit of thermal vacancy formation, irrespective of the crystal orientation. The power α is close to $\frac{1}{2}$, as predicted by the theory, with $\alpha \approx 0.5$ for Mo and $\alpha \approx 0.6$ for Al, Cu, and Ag. No evidence of localized positron states at low temperatures was found.

Various factors having an effect on the exact value of the power α have been discussed. The temperature dependence of the elastic constants seems to account for a part of the deviation from $\alpha = \frac{1}{2}$. The deformation-potential parameters extracted from the measured D_+ values are in quantitative agreement with theoretical calculations, when a value for the positron effective mass $m^* = 1.5m_e$ is used. Combining earlier work-function measurements with our new results yields estimates for the effect of lattice vibrations on the internal electrostatic potential. The effect is found to be significant.

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