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Planar Channeling of Electrons and Positrons in Crystals

Local-Density Calculation of Planar Averaged Potential and Channeling Radiation Spectrum

By

S. SATPATHY[3]) (a) and A. P. PATHAK[4]) (b)

Results of calculation of channeling potentials in crystals using the local density theory are given. The linear muffin-tin orbitals method is used in the atomic spheres approximation (LMTO-ASA) to calculate the planar-averaged potential seen by MeV electrons and positrons channeled along (110) planes of diamond, Si, and Ge. The calculations thus include the spherical charge relaxation effects ("solid state effects") in the solid but omits the non-spherical part. The calculated potentials are in excellent agreement with the empirical potentials derived from channeling radiation experiments, but are significantly different near the atomic planes from the potentials obtained from superposition of the atomic Thomas-Fermi statistical (Molière) potentials. Agreement of the potentials with the ones obtained from superposition of atomic Doyle-Turner potentials is reasonably good. Frequencies of emitted radiation from the channeled electrons and positrons are also calculated and compared to experiments. For the crystals considered, it is concluded that the solid state effects on the planar-averaged potential are moderate to small. It is expected that these effects should be more important for channeling in ionic solids. The deviation of the observed positron channeling radiation from the calculated values for Si and C indicates that the non-spherical part of the charge relaxation might be relatively important for these solids.

Mit der Lokaldichtetheorie werden die Kanalisierungspotentiale in Kristallen berechnet. Lineare Muffin-tin-Orbital-Methoden in der Atomkugelnäherung (LMTO-ASA) werden benutzt, um die planargemittelten Potentiale zu berechnen, die von den MeV-Elektronen und Positronen verspürt werden, die in den (110)-Ebenen von Diamant, Si und Ge kanalisiert werden. Die Berechnungen schließen somit die Kugelladungsrelaxationseffekte („Festkörpereffekte") im Festkörper ein, vernachlässigen jedoch den nichtkugelförmigen Anteil. Die berechneten Potentiale befinden sich in exzellenter Übereinstimmung mit den empirischen Potentialen, die aus Kanalisierungsstrahlungsexperimenten abgeleitet werden, sind jedoch in der Nähe der Atomebenen beträchtlich verschieden von den Potentialen, die aus der Überlagerung der atomaren statistischen Thomas-Fermi (Molière)-Potentiale erhalten werden. Die Übereinstimmung der Potentiale mit denen von der Überlagerung der atomaren Doyle-Turner-Potentiale ist ziemlich gut. Die Frequenzen der Emissionsstrahlung von den kanalisierten Elektronen und Positronen werden ebenfalls berechnet und mit Experimenten verglichen. Für die betrachteten Kristalle wird angenommen, daß die Festkörpereinflüsse auf das planar-gemittelte Potential moderat bis klein sind. Es wird erwartet, daß diese Effekte für Kanalisierung in Ionenfestkörpern wesentlicher sind. Die Abweichung der beobachteten Positronen-Kanalisierungsstrahlung von den berechneten Werten für Si und C zeigt, daß der nichtkugelförmige Anteil der Ladungsrelaxation relativ wichtig für diese Festkörper sein könnte.

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1. Introduction

Since the first observation of radiation from positrons and electrons channeled in silicon single crystals [1, 2], a fair amount of experimental and theoretical effort has been devoted to the subject. The basic phenomenon is a simple and straightforward consequence of partial confinement of charged particle motion in the crystal as was first predicted by Kumakov [3]. The process of steering of charged particles along major crystallographic planes or axes, and hence the confinement of transverse motion for the channeled particle, results in the formation of discrete bound states in the transverse potential. Spontaneous transitions among these bound states are accompanied by emission of observable electromagnetic radiation called channeling radiation. The transverse potential barrier is relatively small so that the particles must be highly directed for channeling to occur. For relativistically fast particles, the combined effects of Lorentz contraction in the longitudinal (forward) direction and the Doppler shift enhances the frequency of radiation in the forward direction by a factor of $2\gamma^2$ $\left(\gamma = (1 - v^2/c^2)^{-1/2}\right)$, so that for MeV electrons and positrons the emitted photon energy is in the keV range.

For planar electron channeling, the confinement of the electron motion in the transverse direction is somewhat analogous to the confinement of electrons or holes in semiconductor quantum wells and superlattices. However, there are important differences. First, the motion of the channeled electron is confined in the transverse direction to a length of the order of 0.5 nm, corresponding to the distance between the crystallographic plane, whereas in a typical superlattice, this length is about 5 to 10 nm. Second, the confinement potential in case of the superlattice is roughly modelled by a square well, while in case of planar channeling, this potential is roughly of an exponential $\exp(-\alpha x)$ type. Finally, while the size of the confinement layer can be easily engineered in the superlattice by varying the thicknesses of the well and the barrier materials, the channel size in the crystal is fixed by the lattice parameters.

The calculation of the spectral features of the channeling radiation involves knowledge of the basic interaction potential between the channeled particle and the atoms of the target crystal. So far almost all such calculations have used superposition of atomic potentials of the Thomas-Fermi type (Molière exponential [4], Lindhard power law [5], or its modifications [6]), or of Doyle-Turner atomic scattering factors derived from relativistic Hartree-Fock atomic fields [7, 8]. None of these potentials include any effect of redistribution of electronic charge in the solid. Even though in covalent solids such as silicon where electrons do not move between atoms such effects are not expected to be large, the solid state effects could be large and must be taken into account in solids where large electron rearrangement takes place. Earlier authors [9] have discussed the question of inclusion of the charge relaxation effects in the channeling potential. The importance of such effects was illustrated by the recent *ab initio* calculations of Barthelat et al. [10] for the Mg–He pair potentials.

In this paper we show that the characteristics of channeling radiation can be predicted from a parameter-free first-principles calculation using the local density approximation to the density functional theory. Our main purpose is to illustrate the applicability of calculations within the framework of the all-electron local density theory to the calculation of channeling potentials and channeling radiations. In spite of the fact that the solid state effects are expected to be small for diamond, Si, and Ge, we have nevertheless chosen to perform our first calculations on these crystals primarily because of the wealth of theoretical and experimental works that already exist on channeling in these materials. Some preliminary results of our calculations have been already reported [11].

2. Planar Averaged Potential

We consider in this paper planar channeling of electrons and positrons in the (110) planes of diamond, Si, and Ge. The channeled particles do not see the charge corrugation of the individual atoms in the channel, rather they experience an average potential for transverse motion. In the forward direction they propagate as more-or-less free particles. The motion in the transverse direction is described by the Schrödinger equation with the relativistic mass [12]

$$\left(-\frac{\hbar^2}{2m}\frac{\mathrm{d}^2}{\mathrm{d}x^2} + V(x) - E\right)\Psi(x) = 0\,. \tag{1}$$

To obtain the planar-averaged potential, $V(x)$, for the transverse motion, we first need to obtain the electronic charge density in the target crystal. This was calculated in a standard manner by solving the Kohn-Sham-Schrödinger local density equations [13] self-consistently using the linear muffin-tin orbital method [14] in the atomic spheres approximation (LMTO-ASA).The von Barth-Hedin potential [15] was used to describe exchange-correlation effects between the electrons in the target crystal.

The channeled electron interacts with the crystal via the electrostatic Coulomb field. This in fact is the entire potential experienced by the channeled electron as the exchange-correlation effects between the channeled electron and those in the crystal are negligible owing to the large kinetic energy of the channeled electron [16]. The exchange-correlation potential varies rather slowly as a function of the electron density ϱ (for instance, the exchange part for the free-electron gas goes as $\varrho^{1/3}$). This being the case, inclusion of the exchange-correlation part of the potential to the Coulomb part would make the planar-averaged potential only about 1.0 eV extra deep at the atomic planes (with respect to the mid-point between the atomic planes) for the case of planar electron channeling along the (110) plane of Si.

In our calculation we obtained the Coulomb part of the potential by solving the Poisson equation in the unit cell of the crystal with the calculated charge density as described earlier. In the atomic spheres approximation (ASA) only the spherically-symmetric part of the potential is retained. Finally, the planar-averaged potential was obtained as a function of distance x from the (110) atomic plane by averaging the Coulomb potential over the plane. The planar-averaged potential thus calculated omits the thermal vibration of the atoms and is valid only for the static lattice. Following the standard procedure used by earlier authors [17, 18], we have taken the vibration of the nuclei into account by modifying the static lattice potential by convoluting it with the amplitude of vibration of the atoms in the crystal corresponding to room temperature. There is no free parameter involved, however. The vibration amplitudes used in our calculations are: for Si 0.0075 nm, for Ge 0.0085 nm, and for C 0.004 nm [19].

Our static lattice as well as the thermally modified potentials for silicon are shown in Fig. 1 along with the empirically derived potential of Pantell and Swent [20]. Furthermore, the Thomas-Fermi-Molière potential is also reproduced from their work. It is clear from this figure that the thermally modified Molière potential is appreciably different from our calculated potential as well as from the empirically-derived potential which was chosen in [20] to fit an expression of the form $C\,\mathrm{e}^{-\alpha x}$ away from the atomic plane and of a simple harmonic oscillator (SHO) type potential close to the plane. The agreement between our potential and the empirically-derived potential is very good apart from small deviations near the atomic planes. Our potential for Si also agrees very well with the one obtained from superposition of the Doyle-Turner (DT) potential. The Doyle-Turner potential is slightly deeper than ours near the

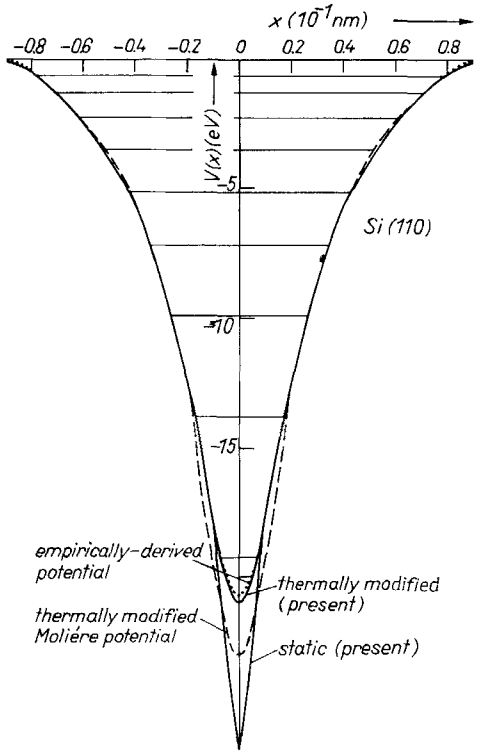

Fig. 1. Planar potentials for electrons channeled along (110) planes of Si. The energy levels correspond to channeling of 54.5 MeV electrons

atomic plane and is slightly shallower away from it: the r.m.s. deviation of the DT potential from ours is only about 0.2 eV, a mere 2% effect.

The calculated planar-averaged potentials for diamond and germanium by the present method are given in Fig. 2 and 3, respectively. For positrons, the planar-

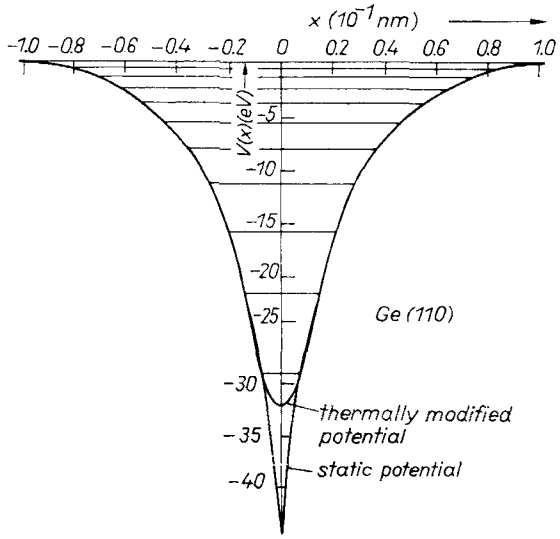

Fig. 2. Planar potentials for electrons channeled along (110) planes of Ge. The energy levels correspond to channeling of 54.2 MeV electrons

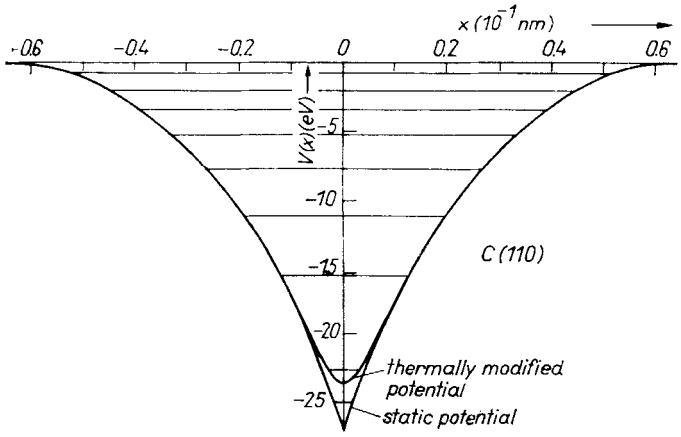

Fig. 3. Planar potentials for electrons channeled along (110) planes of C. The energy levels correspond to channeling of 54.5 MeV electrons

averaged potential is just the negative of that seen by the electron. This potential is shown in Fig. 4 for the case of Si.

3. Channeling Radiation Spectrum

In this section we present the results of our calculation for the characteristics of channeling radiation emitted by electrons and positrons using the channeling potentials calculated in the last section.

3.1 Electron channeling

To calculate the frequency of channeling radiation, we need to solve the Schrödinger equation (1) in the transverse channeling potential. On general grounds [21], the channeling potential may be expected to be of the form

$$V(x) = A + B \exp(-\alpha x) . \tag{2}$$

We find that our calculated potentials for the three crystals fit very well with the expression of the above form. The parameters A, B, α, and the root-mean-square deviation of the fit σ are given in Table 1 for Si, Ge, and diamond.

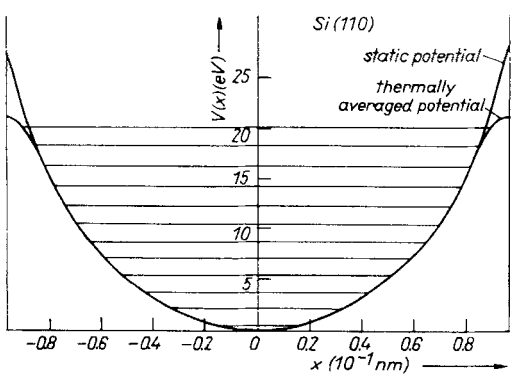

Fig. 4. Planar potentials for positron channeling along (110) planes of Si. The energy levels correspond to channeling of 54.4 MeV positrons.

Table 1

Fitting of the calculated (110) planar-averaged potential with the expression, $V(x) = A + B \exp(-\alpha x)$, for electron channeling in Si, Ge, and diamond. x is the distance from the atomic plane. σ is the r.m.s. deviation of the fit

crystal	lattice constant, a (nm)	A (eV)	B (eV)	α (nm^{-1})	σ (eV)
silicon					
static	0.543	−0.14	−27.1	36.5	0.3
$T = 300$ K	0.543	1.63	−25.0	26.1	0.5
germanium					
static	0.566	−0.82	−44.1	47.8	0.4
$T = 300$ K	0.566	1.36	−37.4	31.9	1.0
diamond					
static	0.356	2.04	−30.3	38.0	0.2
$T = 300$ K	0.356	3.81	−30.2	31.0	0.5

The Schrödinger equation (1) with the potential of the form of (2) is exactly solvable in terms of Bessel functions [21, 20]. In the atomic Rydberg units (unit of length = = Bohr radius, energy unit = Rydberg, and $\hbar^2/2m_e = 1$) and with the relativistic mass $m = \gamma m_e$, the energy eigenvalues are given by

$$E = -\frac{\alpha^2 \nu_i^2}{4\gamma}, \tag{3a}$$

where ν_i's are the zeroes of the Bessel function or its derivative,

$$J_{\nu_i}(\xi_0) = 0 \qquad \text{for odd-parity states,} \tag{3b}$$

$$\left[\frac{dJ_{\nu_i}(\xi)}{d\xi}\right]_{\xi=\xi_0} = 0 \quad \text{for even-parity states} \tag{3c}$$

for a given $\xi_0 \equiv 2(-\gamma B)^{1/2}/\alpha$. The zero of energy has been shifted such that $A = 0$. The eigenfunctions are the Bessel functions $J_{\nu_i}(x)$ of the first kind and order ν_i with the argument $\xi = (2/\alpha)(-\gamma B)^{1/2} \exp(-\alpha x/2)$. We obtained these exact solutions and then used the first-order perturbation theory to correct for the small deviation of the calculated potential from the exponential form. The energy eigenvalues thus obtained for the thermally modified potential and for a channeled electron of specific energy (specific γ value) are sketched in Fig. 1 to 3 along with the planar-averaged potentials.

The emitted photon energy in the forward direction is given by $\hbar\omega = 2\gamma^2(\Delta E)$ where ΔE is the energy difference between the successive initial and final states of the channeled particle. The factor of $2\gamma^2$ comes as mentioned earlier from relativistic effects. The calculated energies of the emitted radiation along with the measured values are given in Table 2. Our results agree well with the experimental values as well as with the Doyle-Turner calculations of Park et al. [22].

3.2 Positron channeling

The transverse potential for positrons is well approximated by a simple harmonic oscillator (SHO) potential. However, the deviation of the calculated potential from the SHO potential is significant and this deviation contributes to the radiation line-

Table 2

Characteristics of (110) planar channeling radiation for electrons in silicon, germanium, and diamond. All energies are in keV

transition	measured peak energy*)	calculated peak energy	
		DT*)	present
silicon (54.5 MeV electrons)			
$1 \rightarrow 0$	122.3 ± 1.0	122.4	126.3
$2 \rightarrow 1$	88.4 ± 0.5	88.9	89.0
$3 \rightarrow 2$	64.2 ± 0.5	64.7	63.8
$4 \rightarrow 3$	49.1 ± 0.6	49.4	48.5
$5 \rightarrow 4$	38.1 ± 0.3	38.4	38.1
$6 \rightarrow 5$	31.6 ± 0.3	29	29.6
diamond (54.5 MeV electrons)			
$1 \rightarrow 0$	161.0 ± 0.5	163.7	164.1
$2 \rightarrow 1$	103.8 ± 0.4	105.7	104.8
$3 \rightarrow 2$	78.0 ± 0.3	79.6	78.5
$4 \rightarrow 3$	60.3 ± 1.5	59.7	58.8
$4 \rightarrow 1$	240.2 ± 1.5	245.0	242.1
germanium (54.2 MeV electrons)			
$1 \rightarrow 0$		176.8	171.7
$2 \rightarrow 1$		138.8	135.2
$3 \rightarrow 2$	**)	105.1	103.1
$4 \rightarrow 3$		78.8	75.1
$5 \rightarrow 4$		59.3	56.8
$6 \rightarrow 5$		44.4	42.9
$7 \rightarrow 6$		32.8	32.5

*) [7] for diamond and [22] for silicon and germanium.
**) Spectral lines in the experiment were not resolved in [22] for germanium.

width. The results of least-squares fitting of the calculated potential with the SHO potential of the form

$$V(x) = V_0 + \tfrac{1}{2} K x^2 \tag{4}$$

is given in Table 3. It should be observed that we have not used (4) as a power-series expansion of the potential $V(x)$ around the minimum at $x = 0$, rather V_0 and K were determined such that our calculated potential is best fitted by the above expression in the least-squares sense in the entire range between the two atomic planes constituting the planar channel. The energies of the quantum levels are equally separated and are given by $E_n = (n + 1/2)\,\hbar\omega_0$ so that the radiation spectrum consists of a single frequency corresponding to transitions between successive energy levels. The deviation from harmonicity causes the harmonic oscillator energy levels to change leading to several lines in the spectrum around ω_0. This, along with other effects, leads to linewidth broadening in the experimental channeling spectrum. In fact, Pantell and Alguard [23] estimate that the anharmonic effects contribute a large part to the linewidth in the positron channeling radiation spectrum.

For calculation of the channeling radiation, we took the exact solutions of the SHO potential and used first-order perturbation theory to take into account the deviation

Table 3

Characteristics of (110) planar channeling radiation for 54.4 MeV positrons at $T = 300$ K. Results of fitting of the calculated positron potential at $T = 300$ K with a simple harmonic oscillator potential of the form $V(x) = V_0 + (1/2)\,Kx^2$ are also given. σ is the standard deviation of the fit

crystal	potential parameters			peak energy (keV)				peak width (keV)	
	V_0 (eV)	K (eV/10^{-2} nm^2)	σ (eV)	exp.	theory			exp.	theory present
					present	Molière	DT		
Si	0.44	46.70	0.63	38.8 ± 0.3	41.4	41.3	41.6	9.5	7.5
Ge	−0.86	65.33	1.82	48.1 ± 0.8	47.6	51.6	52.2	18	18.5
C	1.23	124.00	0.31	65.3 ± 0.3	68.6	63.1	68.5	12	3.0

of the actual potential from the SHO potential. Our calculated radiation frequencies shown in Table 3 are averages of various transitions between successive levels. For calculation of this average, we have taken into account only those transverse states for which the positron motion is confined to the planar channel.

The agreement of the calculated peak energies shown in Table 3 with the experimental values is no better than fair. While the Molière and DT results of earlier authors do not agree very well with experiments for any of the three crystals [22, 24], our calculated peak energy for Ge seems to agree quite well with the experimental value, but for Si and C the agreement is as poor as the DT or the Molière results. Our calculation incorporates only the radial relaxation of the electronic charge caused by the solid formation. This is because the atomic spheres approximation (ASA) in the LMTO theory, which we have employed here, spherically averages all charges about individual atoms in the solid. This approximation might be expected to cause larger errors in the potential in between the atomic planes leading to larger errors for predicted radiation frequencies for positron channeling. This effect should be especially important for crystals with lower atomic numbers, where a larger portion of the electrons are valence electrons. This might explain why our Si and C peak energies do not agree with the experiment as well as our Ge result does.

Our calculated value of the width reported in Table 3 is twice the r.m.s. deviation of various radiation frequencies from the mean. Admittedly, the calculated value of the width of the peak is rather rough as it omits the transition matrix element effects as well as the initial state population distribution of the channeled positron between various eigenstates in the transverse potential, the latter being in fact dependent on the experimental conditions. It is gratifying, however, to note that the calculated width does predict a significant part of the peak width to come from the anharmonic effects consistent with anticipation by earlier authors [23]. The predicted width value of 3.0 keV for diamond is well below the observed value of 12.0 keV consistent with the fact that good crystals of diamond are hard to make. This means that in diamond a large part of the observed linewidth broadening is probably caused by positron dechanneling effects due to crystalline imperfections.

4. Conclusion

In conclusion, we have reported the first calculation of the planar-averaged potential and the channeling radiation spectrum from an *ab initio* local density calculation. Our

calculation of the planar-averaged potential included part of the charge redistribution effect caused by the formation of the solid. Our results are in general in good agreement with the measured channeling radiation characteristics. The discrepancy between our calculated peak energies in positron channeling radiation and the corresponding experimental values in Si and C indicates that non-spherical charge relaxation in these crystals might be important for the positron channeling potential. It would be interesting to go beyond our present approximation and take both the radial as well as then non-spherical charge relaxation into account in the calculation of the positron spectrum.

Our calculations presented here illustrate the applicability of the local density methods to the prediction of channeling radiation spectra. This is an important illustration since the Molière or Doyle-Turner type of potentials that exclude the charge redistribution effects in the solid cannot be expected to be quantitatively reliable especially for ionic solids. A calculation of the type presented here would be in general necessary in such case for obtaining a reasonably accurate channeling potential and hence a reasonable channeling radiation spectrum.

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