tion from the low-temperature region, we estimate that $\mu(\text{expt}) - \mu(k=0) \approx 0.1\ \mu\text{m}$. We hope to have more detailed and precise information on $\mu(k,T)$ from the direct measurement of c_t at the large meander-line wave vector k_L.

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Radiation from Two-Dimensional "Molecular" Bound States of Electrons Channeled in Diamond

J. U. Andersen, S. Datz,[a] E. Lægsgaard, J. P. F. Sellschop,[b] and A. H. Sørensen
Institute of Physics, University of Aarhus, DK-8000 Aarhus C, Denmark
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Axially channeled electrons are captured into bound states of the "atomic-string" potential. When two rows lie in close proximity as in the ⟨110⟩ direction of diamond, the potentials overlap, forming a saddle point between the rows. For 4-MeV electrons in the ⟨110⟩ diamond potential, the $2p$ level lies above the saddle point, and molecular states are formed. The observation of radiation arising from transitions of these states is reported. Spectral information is used to deduce electron-density enhancement in the C-C bond.

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For axial channeling of electrons at a few megaelectronvolts in low-Z materials, transitions between bound states of transverse motion[1] give rise to a line spectrum of "channeling radiation" at energies of a few kiloelectronvolts in the forward (beam) direction.[2,3] In the diamond lattice, the atomic strings along the ⟨110⟩ direction are arranged in pairs, and the bound states of transverse motion have molecular character. In analogy with a diatomic molecule, they may be classified as bonding and antibonding orbitals, and the splitting between these states, which may be derived from the radiation spectra, is particularly sensitive to charge accumulation in the tetrahedral bonds between carbon atoms. The results represent the first clear evidence from channeling radiation of deviations in a crystal from the electron density obtained from overlapping atomic densities. In contrast, for previous experiments on silicon, calculations with atomic scattering factors derived from relativistic Hartree Fock calculations were found to reproduce the measured line energies for channeling radiation within the experimental uncertainty.[3,4]

An electron beam from the 5-MV Van de Graaff

accelerator at Aarhus University was transmitted through a 3.5-μm-thick diamond crystal (type 2A with low defect concentration) mounted in a two-axis goniometer, and magnetically deflected into a Faraday cup. The angular spread of the beam was $\leq 0.02^\circ$. Radiation in the forward direction was measured in a windowless silicon detector with resolution 260 eV at 5.41 keV. The spectra obtained with $\langle 111 \rangle$, $\langle 100 \rangle$, and $\langle 110 \rangle$ directions parallel to the electron beam are shown in Fig. 1. Although there are a number of features present, we are concerned here only with the $2p \rightarrow 1s$ transitions which lie in the 4–6 keV region.

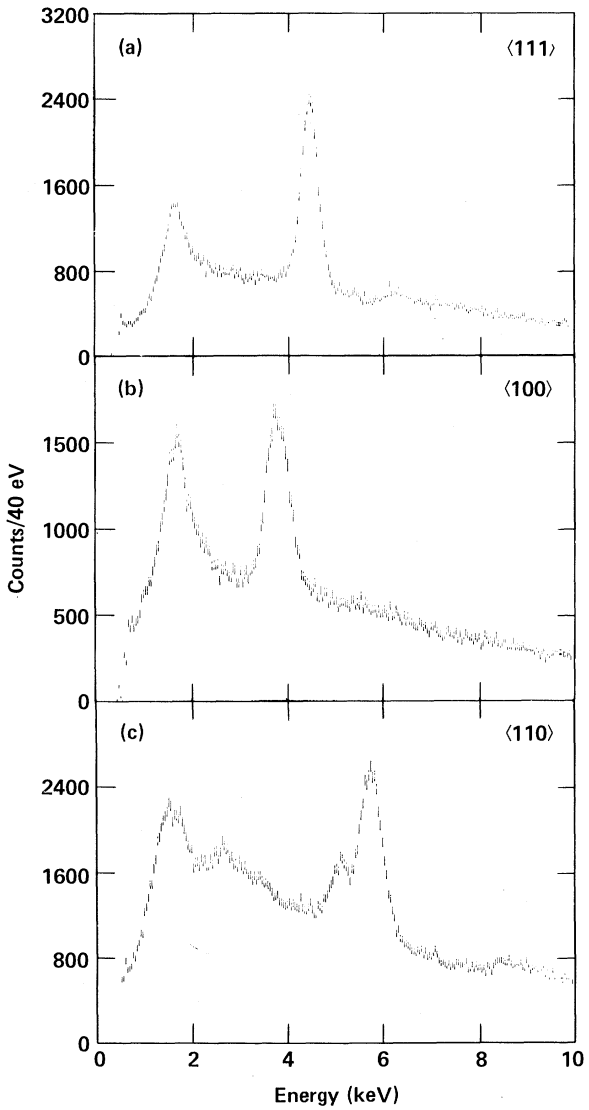

FIG. 1. Spectra of photons generated by 4-MeV electrons channeled along $\langle 111 \rangle$, $\langle 100 \rangle$, and $\langle 110 \rangle$ axes in diamond (3.5 μm thick).

The basic theoretical framework for a description of axial-channeling radiation for electron energies of a few megaelectronvolts was discussed in Ref. 3. The electron wave function is separated into a product of a plane wave describing the motion parallel to the axis and a function depending on the transverse coordinates only. The transverse-energy levels are obtained as eigenvalues of a transverse Hamiltonian corresponding to motion of a particle with the relativistic electron mass $m_0\gamma$ in the two-dimensional, thermally averaged continuum potential.

For channeling radiation in the forward (string) direction, the photon energy is given by

$$\hbar\omega = (1+\beta)\gamma^2 |\Delta E_\perp|, \quad (1)$$

where $\beta = v/c$ and $|\Delta E_\perp|$ is the magnitude of the jump in transverse energy. For isolated states bound to one atomic string, the single-string potential may be applied.[3] The potentials for the $\langle 111 \rangle$ and $\langle 100 \rangle$ directions are qualitatively similar to those shown for silicon in Ref. 3. For the $\langle 111 \rangle$ direction, the $2p$ level lies well below the continuum, and a single sharp line is observed which may be described by a Lorentzian with linewidth $\Gamma = 250$ eV convoluted with the Gaussian detector resolution function. For the weaker $\langle 100 \rangle$ axis, the line is at lower energy and the $2p$ level lies close to the continuum; hence the level is broadened due to band structure.[3] However, the most interesting feature, on which we concentrate in this Letter, is the splitting of the line for the strongest axis, the $\langle 110 \rangle$. A detailed analysis of this spectrum is shown in Fig. 2, and the line energies obtained are given in Table I.

For the $\langle 110 \rangle$ axis (Fig. 3), only the $1s$ level is well described in the single-string approximation since the $2p$ states in a pair of neighboring strings overlap strongly, and the $2p$ level splits into four molecular-type levels. The qualitative features are obtained in a simple treatment analogous to the linear-combination-of-atomic-orbitals (LCAO) method in chemistry. When the transverse Hamiltonian is diagonalized in the subspace spanned by the four single-string $2p$ states for a pair of strings, four eigenstates are obtained which may be classified according to their symmetry under reflection in the midpoint between the strings, gerade and ungerade, and under reflection in the line connecting the two strings, σ and π. The line energies for transitions between molecular energy levels obtained from the LCAO treatment are given in

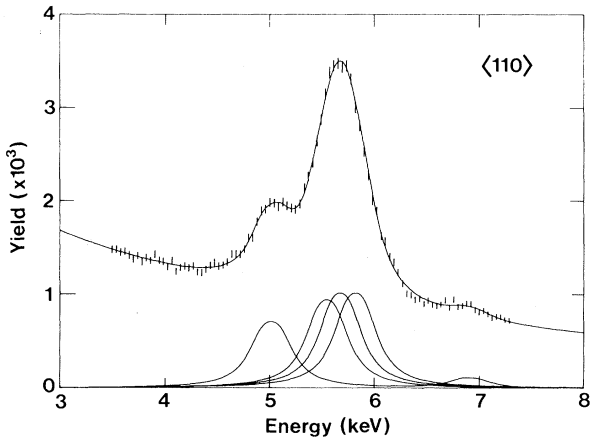

FIG. 2. Decomposition of the ⟨110⟩ $2p$-$1s$ and $2s$-$1s$ lines (see Table I). Fitting of the $2p$-$1s$ lines involved assumptions of equal populations of $2p$ levels, expected because of incoherent scattering, and of equal widths anticipated because of the dominance of the $1s$ state in determining coherence lengths.

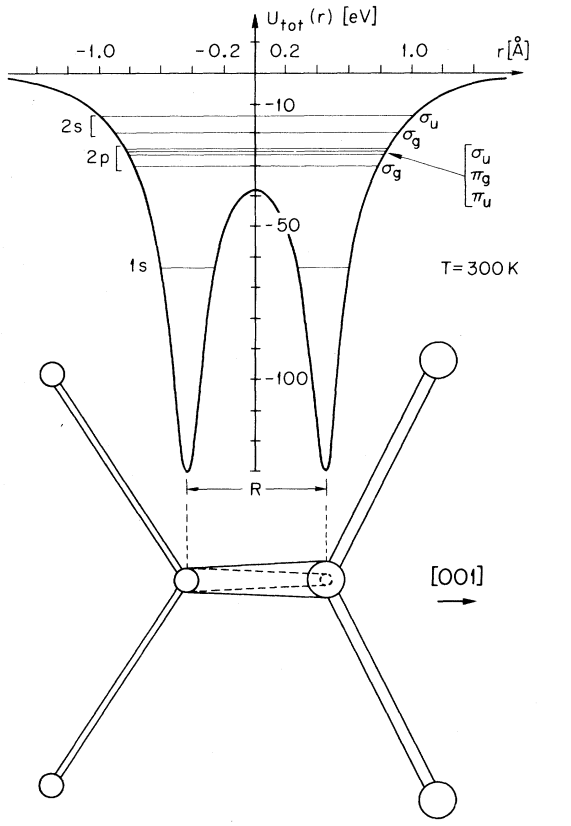

FIG. 3. (Upper) Potential and energy levels for 4-MeV electrons channeled along ⟨110⟩ axis in diamond. (Lower) Atomic core and bond configuration in diamond viewed along the [110] direction.

Table I together with the single-string values. We have also evaluated the mixing with the near-lying $2s$ levels, which turns out to be quite strong, in particular for the $\sigma_g 2p$ level, which is lowered considerably when mixing with the $\sigma_g 2s$ state is introduced. The splitting between the lines is seen to increase by ~150 eV so that it is now ~100 eV larger than the observed splitting. Also, a dipole transition $2s$-$1s$ becomes allowed, and, in fact, a corresponding weak line is visible in the spectrum (see Fig. 2). However, while the LCAO-type model is very instructive for qualitative purposes, its accuracy is limited and difficult to assess.

An alternative and more accurate method involves the many-beam formulation of the Schrödinger equation for the transverse motion.[3] The periodicity of the lattice is used, and the transverse wave function, and the crystal potential averaged in the axial direction and over the

TABLE I. Photon energies (in electronvolts) for $2s$-$1s$ and $2p$-$1s$ transitions for 4-MeV electrons in ⟨110⟩ diamond. The crystal potential was evaluated from Doyle-Turner scattering factors (Ref. 5).

Transition	Single string	1st order LCAO	With $2s$-$2p$ mixing	Many beam	Experiment
$\sigma_g 2s$-$\sigma_u 1s$			7253	7103	6933
$2p$-$1s$	5801				
σ_u-σ_g		6025	6015	6019	5897 [a]
π_g-σ_u		5888		5887	5751 [a]
π_u-σ_g		5771		5742	5624 [a]
σ_g-σ_u		5279	5115	5090	5084 [a]

[a] Corrected for refraction effects (~ 0.6%) and error in beam energy (0.6%).

thermal vibrations, are expanded in Fourier series. The accuracy of such calculations is limited by the number of reciprocal-lattice vectors $\vec{g}$ included. A check is provided by an investigation of the convergence of the result with increasing number of vectors $\vec{g}$ (beams) included. Calculations have been made with up to 361 beams, and the resultant energy levels have then converged to an error of less than 1%. The values obtained for channeling radiation energies from the many-beam formulation are listed in Table I.[6] For the $2p$-$1s$ transitions, there is excellent agreement with the result of the LCAO-type calculations. The upper three lines and the $2s$-$1s$ line are higher than the measured values by ~120 eV. However, the most significant deviation is for the separation of the σ_g-σ_u line, which is larger by ~130 eV than deduced from the experiment (Fig. 2). This we attribute to the accumulation of charge in the tetrahedral bonds in diamond.

The only experimental information on the bond charge in diamond comes from x-ray diffraction measurements, and the data have been analyzed by a number of authors, using various methods of representing density distributions.[7] We have made a simple analysis, in which the bond-charge density is represented by Gaussian distributions centered in the middle of the bonds and containing n electrons per atom. The atomic L-shell density is renormalized through multiplication by $1-n/4$. With Gaussian widths perpendicular to the bond $\sigma_\perp = d_0/10.5$ and parallel to the bond $\sigma_\parallel = d_0/8.5$ a value of $n = 1.7$ gives an excellent fit to the x-ray data ($d_0 = 3.56$ Å, the lattice constant).

The effect on the $\langle 110 \rangle$ potential of a charge accumulation in the bond regions may be understood qualitatively from Fig. 3. Each atom in a $\langle 110 \rangle$ row is bonded to two atoms in a neighboring row and to one atom in each of the next-nearest neighboring rows. Hence accumulation of charge in the bonds increases the electron density between close-lying pairs of $\langle 110 \rangle$ rows and increases the potential energy of channeled electrons in this region. Since the $\sigma_g 2p$ state has a high density here, this level will increase in energy relative to that of other $2p$ levels, i.e., the splitting will be reduced, as required by the experiment. With use of the many-beam method and the Gaussian parameters obtained from the x-ray analysis, the energy levels have been recalculated as a function of n. The values obtained are almost linear in the n region in question, and the closest match with the observed splitting is obtained with $n = 1.45$, corresponding to an electron density of $1.7/\text{Å}^3$ in the center of bonds. Although this determination is probably less accurate than that obtained from the x-ray data, it does demonstrate that information on charge distributions and potentials in crystals can be obtained from channeling radiation.

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[a]Permanent address: Oak Ridge National Laboratory, Oak Ridge, Tenn. 37830.

[b]Permanent address: Department of Physics, University of Witwatersrand, Johannesburg, South Africa.

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