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Emission of Charged Particles from Crystals

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A quantum-mechanical treatment of the problem of the charged-particle emission from crystals has been presented. In addition to the mass and potential dependence established by the previous wave-mechanical calculations, we have illustrated the energy and the temperature dependence by assuming the crystal to be initially in a low-lying state and calculating the renormalization of the particle wave function using the Debye model for the lattice vibrations. The attenuation is shown to be a natural consequence of the inelastic processes, and its magnitude is found to be small compared to previous conjectures.

I. INTRODUCTION

Ever since Lindhard[1] proposed the theoretical basis for the channeling of particles in perfect crystals based on the ideas of the strings of atoms and the critical angle, a good deal of experimental work has been done,[2-5] mostly on the channeling of heavy ions in perfect crystals. There has been excellent agreement between experimental and theoretical values of the critical angle, the range of the ions in the crystal, and the other parameters. However, for light particles such as electrons and positrons, Lindhard's classical treatment has been shown[6,7] to give only the gross features of the phonomena, and it has been found in a quantum-mechanical treatment that the phenomena must show a mass dependence, in contradiction to the classical predictions. Actually, similar deviations from classical results had been pointed out by Lervig *et al.*,[8] who showed that for electrons and positrons the penetration into the classically forbidden region will be significant and the quantum effects must be taken into account. A similar quantum-mechanical treatment of the electron and positron channeling has been proposed by Howie.[9]

The experimental situation for light-particle channeling seems to be poor. A recent communication[10] gives some information of electron channeling at high energies. However, the charged-particle emission problems, which are governed by the same physical mechanism as the channeling problems, have attracted experimental interest. A significant work in this connection is that of Uggerhøj[11] on the electron and positron emission from ^{64}Cu embedded in a copper crystal. Very recently Walker *et al.*[12] have studied the effects of bremsstrahlung radiations on the channeling of relativistic electrons and positrons.

In order to make some comparison with the experiments of Uggerhøj, DeWames *et al.*[6,7] have quantum mechanically treated the emission of electrons and positrons from crystals. Using the periodic potential field $U(r)$ of the classical model in an approximate form[13] of the many-particle Schrödinger equation, they have shown how the quantum-mechanical treatment displays mass and potential dependence of the charged-particle emission from crystals. Their results completely dispose, once and for all, of the notion that the emitted intensity pattern for charged particles, regardless of mass, consists of a broad dominant envelope on top of which is superposed small detailed structure due to the (quantum-mechanical) Bragg resonances. However, the inelastic processes such as emission and absorption of the phonons by the emitted particle have been ignored in this formalism. These inelastic processes have been found[14] to lead to the renormalization of the particle wave function which is of basic importance in the theory of anomalous transmission of particles through perfect crystals.[15]

In this paper we consider the propagation of particles through crystals. Starting with the Schrödinger equation for the whole system (crystal and particle), we utilize the standard Born approximation[14] to solve coupled particle-crystal equations for the amplitude of the transmitted wave. Then the reciprocity relation[6,9,13] is used to relate the problems of emission and penetration of particles. Assuming the crystal to be initially in a low-lying state, the formulation has been given in Sec. II and the Debye model for the lattice vibrations has been used to evaluate the renormalization matrix elements in Sec. III. The conclusions regarding the temperature and energy dependence of the process, in addition to those of mass and potential dependence, have been summarized in Sec. IV. The numerical values of the absorption parameter calculated from the present formalism differ appreciably from those obtained in earlier diffraction theories.

II. FORMULATION AND RENORMALIZATION MATRIX ELEMENTS

We consider the emission of particles from a source embedded in a perfect crystal. The crystal is assumed to be initially in a low-lying state $|n\rangle$ at low temperature. Leaving aside the actual mechanism of emission, we shall be concerned only with the propagation of the emitted particle through the crystal and its subsequent escape from the crystal. We shall also neglect the interactions (if any) of the emitted particle with the emitter itself, and thus we are ignoring the processes that might occur in the immediate vicinity of the emitter. Under these simplifying assumptions we are faced with the problem of propagation of particles through the periodic potential field of the crystal.

The Schrödinger equation describing our system (the crystal and the particle) is

$$H\Psi = E\Psi \ , \tag{1}$$

where the total Hamiltonian H is given by $H = H_0 + H_p + V$, H_0 being the crystal Hamiltonian, H_p the free-particle Hamiltonian, and V the interaction potential between the particle and the crystal. The total energy of the system E is given by $E = E_n + E_p$, E_n being the energy of the crystal when it is in the nth phonon state $(H_0|n\rangle = E_n|n\rangle)$, and E_p is the particle energy at emission. The total wave function of the system Ψ can be expanded[14] in terms of the crystal eigenfunctions $|n\rangle$ corresponding to energy E_n:

$$\Psi(\vec{r}, \{\vec{R}_\sigma\}) = \textstyle\sum_n |n\rangle\, \varphi_n(\vec{r}) \ , \tag{2}$$

where $\vec{r}$ is the position of the particle and $\vec{R}_\sigma$ is the actual position of σth nucleus in the crystal. Using (2) in (1), we may write the equations coupling the particle and the crystal wave functions in the form[16]

$$\left(-\frac{\hbar^2\nabla^2}{2m_0} + \langle n|V|n\rangle - E_p\right)\varphi_n(\vec{r}) = -\sum_{m\neq n} \langle n|V|m\rangle\, \varphi_m(\vec{r}) \ , \tag{3}$$

where m_0 is the mass of the particle. The right-hand side of Eq. (3) represents the renormalization of the particle wave function (due to inelastic processes) which is missing in the calculations of DeWames *et al.*[6,7] The intensity at a point $\vec{r}$ outside the crystal due to emission at a point $\vec{r}_e$ inside the crystal is obtained by calculating the intensity at the emitter has been placed at the observation point $\vec{r}$. This can be done in view of the reciprocity relation[13]

$$\varphi_n(\vec{r}, \vec{r}_e) = \varphi_n(\vec{r}_e, \vec{r}) \ , \tag{4}$$

which holds[17] as long as one neglects the reflections at the crystal surface. Thus the problem is reduced to that of finding the intensity at the emitter site, when the particles are coming from a faraway point $\vec{r}$.

The problem of penetration of particles has been treated in detail in an earlier work[15] (hereafter referred to as I). Using the Born approximation[14] for $\varphi_m(\vec{r})$ appearing on right-hand side of (3), we write (3) as in I:

$$(2\delta + \zeta_h) u_h(n) + \textstyle\sum_g \psi_{h-g} u_g(n) = 0 \ , \tag{5}$$

where the expansions[6,7,14,15]

$$\varphi_n(\vec{r}) = \textstyle\sum_h u_h(n)\, e^{i(\vec{k}_M + \vec{K}_h)\cdot\vec{r}} \tag{6}$$

and

$$V_{nn}(\vec{r}) = \textstyle\sum_h V_h(n)\, e^{i\vec{K}_h\cdot\vec{r}}$$

have been used. In Eq. (5),

$$\begin{aligned} 2\delta E_p &= (\hbar^2/2m_0)\, k_M^2 - E_p \ , \\ (\hbar^2/2m_0)(K_h^2 + 2\vec{K}_h\cdot\vec{k}_M) &= \zeta_h E_p \ , \\ \psi_{h-g} &= [V_{h-g} + C_{hg}(n)]/E_p \ , \end{aligned} \tag{7}$$

with the renormalization matrix elements given by

$$C_{hg}(n) = -\frac{2m_0}{V'\hbar^2}\int d\vec{r}\int d\vec{r}'\, e^{-i(\vec{K}_h + \vec{k}_n)\cdot\vec{r} + i(\vec{K}_g + \vec{k}_n)\cdot\vec{r}'} \times \sum_{n'\neq n} V_{nn'}(\vec{r})\,\frac{e^{ik_n'|\vec{r}-\vec{r}'|}}{4\pi|\vec{r}-\vec{r}'|} \ , \tag{8}$$

where $k_n^2 = 2m_0E_p/\hbar^2$ and k_M is approximately equal to k_n, with a small imaginary part (I). Equation (5) is similar to Eq. (7) of De.Wames *et al.*[7] but the renormalization of the wave function has been taken into account which has the effect of adding $C_{hg}(n)$ to $V_{h-g}(n)$. The significance of this is that one need not take a phenomenological attenuation parameter in the form of the imaginary part of the potential. The relevant quantity is supplied here by $\mathrm{Im}C_{hg}$.

Using the proper boundary conditions on the wave function at the surface of the crystal, we get the final equation for the intensity in two-beam theory (single Bragg reflection), when the emitter is located at a lattice site[6]:

$$\begin{aligned} |\varphi_n(t)|^2 = {} & x^2 \exp\{-[1-\epsilon_h(1+y^2)^{-1/2}]t/\xi_0''\} \\ & + (1-x)^2 \exp\{-[1+\epsilon_h(1+y^2)^{-1/2}]t/\xi_0''\} \\ & + 2x(1-x)\exp(-t/\xi_0'')\cos[(1+y)^{1/2}t/\xi_h'] \ , \end{aligned} \tag{9}$$

where

$$\begin{aligned} x &= \tfrac{1}{2}[1 + (y-1)(1+y^2)^{-1/2}], \\ y &= \zeta_h/2\psi_h' \quad (\mathrm{Re}\psi_h = \psi_h' = [V_h + \mathrm{Re}C_{h0}]/E_p) \ , \\ \epsilon_h &= \psi_h''/\psi_0'' \quad (\mathrm{Im}\psi_h = \psi_h'' = \mathrm{Im}C_{h0}/E_p) \ , \\ \xi_h' &= (k_0\psi_h')^{-1} \ , \quad \xi_h'' = -(k_0\psi_h'')^{-1} \ , \end{aligned} \tag{10}$$

and t is the distance of the emitting atom from the crystal surface. In deriving (9), the assumption of $\psi_h'' = \mathrm{Im}C_{h0}/E_p$ being small compared to ψ_h' is essential and this assumption will be justified when we calculate the renormalization matrix elements.

The imaginary part of the renormalization matrix has been calculated in I. We follow exactly the same method and similar approximations for evaluation of the real part. From Eq. (8) we have

$$\mathrm{Re}C_{hg}(n) = -\frac{2m_D}{V'\hbar^2(2\pi)^3}\int d\vec{k}\int d\vec{r}\int d\vec{r}'\, e^{i(\vec{k}-\vec{k}_n-\vec{K}_h)\cdot\vec{r} - i(\vec{k}-\vec{k}_n-\vec{K}_g)\circ\vec{r}'} \times \sum_{n'\neq n} V_{nn'}(\vec{r})\, V_{n'n}(\vec{r}')\, P\!\left(\frac{1}{k^2 - k_{n'}^2}\right) \ , \tag{11}$$

where $P(1/x)$ represents the principal value. Now we take a general interaction potential $V(\vec{r}) = \sum_\sigma V_\sigma(\vec{r} - \vec{R}_\sigma)$, $V_\sigma(\vec{r} - \vec{R}_\sigma)$ representing the interaction of the particle with the σth atom of the crystal. Calculating the matrix elements involved in the one-phonon approximation[18] assuming the crystal to have no isotopes and zero nuclear spin as in I, we get

$$\begin{aligned} \mathrm{Re}C_{hg}(n) = -\frac{m_0}{Mv_c(2\pi)^3}\int\frac{d\vec{f}}{\xi}\Bigg[& \frac{V(\vec{f}+\vec{K}_h)\,V(\vec{f}+\vec{K}_g)}{1-e^{-\xi/k_BT}}\,(\vec{f}+\vec{K}_h)\circ(\vec{f}+\vec{K}_g)\; e^{-D[(\vec{f}+\vec{K}_h)^2+(\vec{f}+\vec{K}_g)^2]}P\!\left(f^2 - 2\vec{f}\cdot\vec{k}_n + \frac{2m_0\xi}{\hbar^2}\right)^{-1} \\ & + \frac{V(\vec{f}-\vec{K}_h)\,V(\vec{f}-\vec{K}_g)}{e^{\xi/k_BT}-1}\,(\vec{f}-\vec{K}_h)\cdot(\vec{f}-\vec{K}_g)\; e^{-D[(\vec{f}-\vec{K}_h)^2+(\vec{f}-\vec{K}_g)^2]}P\!\left(f^2 + 2\vec{f}\cdot\vec{k}_n - \frac{2m_0\xi}{\hbar^2}\right)^{-1}\Bigg] \ , \end{aligned} \tag{12}$$

with

$$D = \left(\frac{\hbar^2}{4MN}\right)\sum_j \frac{\coth(\xi_j/2k_BT)}{\xi_j} \ ,$$

where $V(\vec{K}) = V_\sigma(\vec{K}) = \int V_\sigma(\vec{r})e^{i\vec{K}\cdot\vec{r}}\,d\vec{r}$; ξ and $\vec{f}$ are, respectively, the energy and the wave vector of the phonon exchanged. We have summed (in the sense of averaging) over all possible values of f to include the possibility of any phonon being exchanged. M is the atomic mass of the crystal atoms, and v_c is the volume of the unit cell.

This will now be calculated in the Debye model choosing screened Coulomb potential as the interaction between the particle and the crystal.

III. RESULTS FOR DEBYE MODEL AND SCREENED COULOMB POTENTIAL

An important approximation for the phonon dispersion is to use the Debye model which correponds to the low-frequency part of any actual dispersion. Thus we set $\xi = \hbar c f$, where c is the velocity of the acoustic vibrations in the crystal and the maximum value of f can be f_0.

For charged particles, one can choose the interaction as in I,

$$V_\sigma(\vec{r}-\vec{R}_\sigma) = z_1 z_2 e^2 \frac{e^{-\Lambda|\vec{r}-\vec{R}_\sigma|}}{|\vec{r}-\vec{R}_\sigma|} , \tag{13}$$

where Λ is the screening parameter, z_1 is the charge of the incident particle in units of e, and z_2 is the atomic number of the crystal atoms. For this interaction we have

$$V(\vec{K}) = V_\sigma(\vec{K}) = \frac{4\pi z_1 z_2 e^2}{\Lambda^2 + K^2} ,$$

so that Eq. (12) becomes

$$\operatorname{Re}C_{hg}(n) = -\frac{2m_0(z_1z_2e^2)^2}{\pi M\hbar c v_c}\int\frac{d\vec{f}}{f}\left[\frac{(\vec{f}+\vec{K}_h)\cdot(\vec{f}+\vec{K}_g)}{1-e^{-\hbar cf/k_BT}}\,P\left(f^2-2\vec{f}\cdot\vec{k}_n+\frac{2m_0cf}{\hbar}\right)^{-1}\frac{e^{-D[(\vec{f}+\vec{K}_h)^2+(\vec{f}+\vec{K}_g)^2]}}{[\Lambda^2+(\vec{f}+\vec{K}_h)^2][\Lambda^2+(\vec{f}+\vec{K}_g)^2]}\right.$$
$$\left.+\frac{(\vec{f}-\vec{K}_h)\cdot(\vec{f}-\vec{K}_g)}{e^{\hbar cf/k_BT}-1}\,P\left(f^2+2\vec{f}\cdot\vec{k}_n-\frac{2m_0cf}{\hbar}\right)^{-1}\frac{e^{-D[(\vec{f}-\vec{K}_h)^2+(\vec{f}-\vec{K}_g)^2]}}{[\Lambda^2+(\vec{f}-\vec{K}_h)^2][\Lambda^2+(\vec{f}-\vec{K}_g)^2]}\right] . \tag{14}$$

Since the upper limit of the f integration, f_0, is always small compared to a reciprocal-lattice vector, we can neglect f compared to K_h as in I, to get

$$\operatorname{Re}C_{h0} = -\frac{2m_0(z_1z_2e^2)^2\,e^{-DK_h^2}\rho}{\pi M\hbar c(\Lambda^2+K_h^2)}\int\frac{d\vec{f}}{f}\left[\frac{\vec{f}\cdot(\vec{f}+\vec{K}_h)}{1+e^{-\hbar cf/k_BT}}\,P\left(f^2-2\vec{f}\cdot\vec{k}_n+\frac{2m_0cf}{\hbar}\right)^{-1}\frac{1}{\Lambda^2+f^2}\right.$$
$$\left.+\frac{\vec{f}\cdot(\vec{f}-\vec{K}_h)}{e^{\hbar cf/k_BT}-1}\,P\left(f^2+2\vec{f}\circ\vec{k}_n-\frac{2m_0cf}{\hbar}\right)^{-1}\frac{1}{\Lambda^2+f^2}\right] , \tag{15}$$

where $\rho = 1/v_c$ is the number density of atoms in the crystal. The angular integration involved in Eq. (15) may be done by choosing k_n as Z axis as in I. The results under the approximation of neglecting m_0c compared to $\hbar k_n$ (which is justified for electrons with energy of the order keV or higher) yields

$$\operatorname{Re}C_{h0} = -\frac{2m_0(z_1z_2e^2)^2\rho\,e^{-DK_h^2}}{M\hbar ck_n(\Lambda^2+K_h^2)}\int_0^{f_0}\frac{fdf}{\Lambda^2+f^2}\left\{\left[f\left(1+\frac{K_h\cos\alpha}{2k_n}\right)\coth\left(\frac{\hbar cf}{2k_BT}\right)+\frac{m_0cK_h\cos\alpha}{\hbar k_n}\right]\ln\frac{2k_n+f}{2k_n-f}\right.$$
$$\left.-2K_h\cos\alpha\coth\left(\frac{\hbar cf}{2k_BT}\right)\right\} , \tag{16}$$

where α is complementary to the Bragg angle. The integration in Eq. (16) may be carried out numerically. In order to get an estimate, we simplify the situation by taking $\ln(1+f/2k_n)\simeq f/2k_n$, since f_0 is small compared to k_n, restricting to low temperatures so that $\coth(\hbar cf/2k_BT)\simeq 1+2e^{-\hbar cf/k_BT}$. Thus we get

$$\operatorname{Re}C_{h0} = -\frac{2m_0\rho(z_1z_2e^2)^2e^{-D_lK_h^2}}{M\hbar ck_n^2(\Lambda^2+K_h^2)}\left\{\frac{1}{2}\left(1+\frac{K_h\cos\alpha}{2k_n}\right)\left[f_0^2-\Lambda^2\ln\left(1+\frac{f_0^2}{\Lambda^2}\right)\right]+\frac{m_0cK_h\cos\alpha}{\hbar k_n}\left(f_0-\Lambda\tan^{-1}\frac{f_0}{\Lambda}\right)\right.$$
$$+2\left(1+\frac{K_h\cos\alpha}{2k_n}\right)f_0^2\left[(1-e^{-\Theta_D/T})\left(\frac{T}{\Theta_D}\right)^2-\frac{T}{\Theta_D}e^{-\Theta_D/T}\right]-K_hk_n\cos\alpha\ln\left(1+\frac{f_0^2}{\Lambda^2}\right)$$
$$\left.+2\left[\left(1+\frac{K_h\cos\alpha}{2k_n}\right)^2\Lambda^2+2K_hk_n\cos\alpha\right]\left[\operatorname{ci}\left(\frac{\Lambda\hbar C}{k_BT}\right)\cos\left(\frac{\Lambda\hbar C}{k_BT}\right)+\operatorname{si}\left(\frac{\Lambda\hbar C}{k_BT}\right)\sin\left(\frac{\Lambda\hbar C}{k_BT}\right)\right]\right\} . \tag{17}$$

Now from the Bragg condition, $K_h\cos\alpha/2k_n = -(K_h/2k_n)^2$, so that if one neglects $(K_h/2k_n)^2$ compared to unity (for high-energy particles) then one gets finally

$$\operatorname{Re}C_{h0} = -\frac{\hbar\rho(z_1z_2e^2)^2e^{-D_lK_h^2}}{Mc(\Lambda^2+K_h^2)E_p}\left\{\tfrac{1}{2}f_0^2\left[1-\frac{\Lambda^2}{f_0^2}\ln\left(1+\frac{f_0^2}{\Lambda^2}\right)\right]+2f_0^2\left[\left(\frac{T}{\Theta_D}\right)^2(1-e^{-\Theta_D/T})-\frac{T}{\Theta_D}e^{-\Theta_D/T}\right]\right.$$

$$+\tfrac{1}{2}K_h^2\ln\left(1+\frac{f_0^2}{\Lambda^2}\right)+2(\Lambda^2-K_h^2)\left[\mathrm{ci}\left(\frac{\Lambda\hbar C}{k_BT}\right)\cos\left(\frac{\Lambda\hbar C}{k_BT}\right)+\mathrm{si}\left(\frac{\Lambda\hbar C}{k_BT}\right)\sin\left(\frac{\Lambda\hbar C}{k_BT}\right)\right]\Bigg\}. \qquad (18)$$

Using the same approximations, Eq. (39) of I gives

$$\mathrm{Im}C_{h0}=-\frac{\pi\rho(2m_0)^{1/2}(z_1z_2e^2)^2e^{-D_lK_h^2}}{MCE_p^{1/2}(\Lambda^2+K_h^2)}F_0(\Lambda,T), \qquad (19)$$

with

$$F_0(\Lambda,T)=f_0+2f_0\left(\frac{T}{\Theta_D}\right)(1-e^{-\Theta_D/T})-\Lambda\left[\tan^{-1}\frac{f_0}{\Lambda}+\mathrm{ci}\left(\frac{\Lambda\hbar c}{k_BT}\right)\sin\left(\frac{\Lambda\hbar c}{k_BT}\right)-\mathrm{si}\left(\frac{\Lambda\hbar c}{k_BT}\right)\right]. \qquad (20)$$

The Fourier transform of the initial state expectation value of the interaction potential $\langle n|V(\vec{r})|n\rangle$ is given by

$$V_h=\frac{1}{V'}\int d\vec{r}\,\langle n|V(\vec{r})|n\rangle e^{-i\vec{K}_h\cdot\vec{r}}$$

$$=\frac{V(K_h)}{V'}\sum_{\vec{\sigma}}\langle n|e^{-i\vec{K}_h\cdot\vec{u}_\sigma}|n\rangle .$$

Evaluating the matrix elements and averaging over the initial states as in Kothari and Singwi,[18] one gets the temperature dependence of V_h (and hence of intensity) through the Debye-Waller factor

$$V_h=\frac{4\pi z_1z_2e^2}{(\Lambda^2+K_h^2)}\rho e^{-W}, \qquad (21)$$

with

$$W=\frac{\hbar^2}{4MN}K_h^2\sum_j\frac{\coth(\xi_j/2k_BT)}{\xi_j}$$

$$=\frac{3\hbar^2}{8k_B\Theta_DM}\left[1+4\left(\frac{T}{\Theta_D}\right)^2\right]K_h^2$$

(in the low-temperature limit). (22)

The expressions (18) and (21) for $\mathrm{Re}C_{h0}$ and V_h are to be employed for the calculation of the emitted intensity from Eqs. (9) and (10). It has been found on numerical evaluation that for $\Lambda=4.2\times10^8\ \mathrm{cm}^{-1}$, $k_n=2\times10^{10}\ \mathrm{cm}^{-1}$, $|V_h/E_p|=10^{-4}$ (the values taken by DeWames *et al.*[6]) corresponding to the copper crystal, the magnitude of $\mathrm{Re}C_{h0}/V_h$ is about 10^{-5} and the magnitude of $\mathrm{Im}C_{h0}/V_h$ is about 10^{-3}. Thus the resulting angular distribution patterns of the emitted intensity will be almost the same as those of DeWames *et al.*[6,7] without attenuation. However, their curves (Figs. 5 and 9 of Ref. 6) correspond to an overestimation of the attenuation ($\psi_h''/\psi_h'\sim0.1$) in view of the fact that, from above, $\mathrm{Im}C_{h0}/V_h\sim10^{-3}$, implying very little attenuation. Indeed, ψ_h'' in our calculation is determined once the parameters for ψ_h' are fixed. Thus the renormalization of the particle wave function takes care of the attenuation which is now proportional to $(m_0/E_p)^{1/2}$, implying merely that for keV electrons and positrons $\mathrm{Im}C_{h0}/V_h\sim10^{-2}$. Since $\mathrm{Re}C_{h0}$ is inversely proportional to E_p (and is independent of the particle mass), for keV particles $\mathrm{Re}C_{h0}/V_h\sim10^{-3}$. We note that the present results for the absorption parameter $\mathrm{Im}C_{h0}/V_h$ differ from the results for V_h'/V_h (contribution from phonon excitations) in earlier electron diffraction theories[19,20] (V_h' is the imaginary part of the Fourier transform of the complex interaction potential and plays the same role as $\mathrm{Im}C_{h0}$ in our calculations). One might doubt here that the use of one-phonon processes with the Debye model for lattice vibrations in the calculation of $\mathrm{Re}C_{h0}$ and $\mathrm{Im}C_{h0}$ is responsible for this difference. Then it is natural to look for the contribution to $\mathrm{Im}C_{h0}$ from the multiphonon processes which, however, has been shown[15,18] to be small compared to the contribution from one-phonon process. On the other hand, as Hall and Hirsch[20] have pointed out, the Einstein's model may be more appropriate than the one-phonon Debye model[21] in view of the accurate results obtained from it for total inelastic scattering cross section. To this end, we note that if we use the expressions for $\mathrm{Re}C_{h0}$ and $\mathrm{Im}C_{h0}$ in the Einstein model calculated by DeWames *et al.*,[14] the values of $\mathrm{Re}C_{h0}/V_h$ and $\mathrm{Im}C_{h0}/V_h$ come out to be of the same order of magnitude as obtained here in the Debye model.

Another interesting feature of the present formalism is that $\mathrm{Re}C_{h0}$ and $\mathrm{Im}C_{h0}$ do not depend on the sign of the charge of the particle. Therefore the ratio $\kappa=\mathrm{Im}C_{h0}/V_h$ is positive for electrons and negative for positrons. Then Eqs. (10) show that the scale y is approximately given by[6]

$$y=\frac{\sin2\theta_B}{(V_h+\mathrm{Re}C_{h0})/E_p}(\theta_B-\theta). \qquad (23)$$

It follows that the magnitude of y for $\theta=0$ is slightly increased for the positrons and slightly decreased for the electrons (since $\mathrm{Re}C_{h0}$ is always negative). This shows qualitatively that the angular width of the intensity pattern must be smaller in the case of the electrons as compared to that of the positron. This feature which arises from the renormalization of the particle wave function is absent in the usual two-beam theories.[10,20]

Now from Eq. (23) we see that for no attenuation, the thickness-averaged intensity of the two-beam solution (9) has the outer wing in the intensity pattern at $y = \pm 1$:

$$\theta_w = \theta_B + \Delta\theta_B$$

$$= \frac{1}{2}\left(\frac{\hbar K_h}{(2m_0 E_p)^{1/2}}\right) + \frac{|V_h| + |\mathrm{Re}C_{h0}|}{\hbar K_h}\left(\frac{2m_0}{E_p}\right)^{1/2} \quad \text{(for electrons)} \quad (24)$$

$$= \frac{1}{2}\left(\frac{\hbar K_h}{(2m_0 E_p)^{1/2}}\right) + \frac{|V_h| - |\mathrm{Re}C_{h0}|}{\hbar K_h}\left(\frac{2m_0}{E_p}\right)^{1/2} \quad \text{(for positrons).} \quad (25)$$

This shows that the condition $\Delta\theta_B/\theta_B \ll 1$ for validity of the two-beam theory is energy dependent (through $\mathrm{Re}C_{h0}$) and is more favorable for the positrons than for the electrons at lower energies where $\mathrm{Re}C_{h0}$ starts contributing. At these low energies, the angular characteristics are also expected to change because the energy is no longer simply a scaling factor.

IV. CONCLUSION

From the present calculations and discussions, some important conclusions regarding the charged-particle emission from crystals emerge. The fact that the magnitudes of $\mathrm{Re}C_{h0}$ and $\mathrm{Im}C_{h0}$ are small compared to the Fourier transform of the potential, shows that the renormalization of the wave function does not change the intensity patterns appreciably and it also indicates that at higher energies the inelastic processes such as phonon emission and absorption do not participate very significantly in channeling of electrons and positrons into crystals. At low energies (of the order of keV or below) $\mathrm{Re}C_{h0}$ starts increasing as $1/E_p$ and then the importance of more accurate treatment such as presented here becomes apparent. As far as the quantitative results are concerned, we note that even when the renormalization of the wave function is neglected, the periodic crystal potential field $U(r)$ of the classical model should be replaced by the expectation value of the actual interaction potential in the initial state of the crystal. This naturally takes account of the lattice vibrations, and when one takes an average over initial states, one gets the temperature dependence of the process through the Debye-Waller factor.

The attenuation of the emitted particle in the crystal has usually been[6,7] treated by assuming an imaginary part of the interaction potential as in electron diffraction theories.[19,20] Although DeWames *et al.*[6] have chosen the value for the ratio of imaginary to real part as approximately 0.1, the detailed variation of this quantity with the magnitude of the reciprocal-lattice vector and the temperature have been given by Humphreys and Hirsch.[19] In the present formalism, the corresponding quantity is supplied by $\mathrm{Im}C_{h0}/V_h$ and is a natural consequence of renormalization of the particle wave function due to inelastic processes. The value of $\mathrm{Im}C_{h0}/V_h$ has been found to disagree with V_h'/V_h obtained in the above-mentioned diffraction theories and is small, showing that absorption is small in the present formalism. The dependence of $\mathrm{Im}C_{h0}/V_0$ on the mass and energy of the incident particle as $(m_0/E_p)^{1/2}$ shows that attenuation for heavy particles at lower energies will be larger. Finally, it is interesting to note that the present formalism qualitatively shows a difference between the widths of electron and positron emission patterns even in the two-beam theory.

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Nuclear Magnetic Resonance and Relaxation in the "Liquid Semiconductors" In_2Te_3, Ga_2Te_3, and Sb_2Te_3

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Measurements of the Knight shifts of In^{115}, Ga^{69}, Ga^{71}, Sb^{121}, Sb^{123}, and Te^{125} in solid and liquid In_2Te_3, Ga_2Te_3, and Sb_2Te_3 are reported for temperatures ranging from roughly 150 K below the melting points to 1400–1500 K. Measurements of the nuclear spin-spin relaxation rates of In^{115}, Ga^{69}, Ga^{71}, Sb^{121}, and Sb^{123} are reported for the liquids from the melting points to 1400–1500 K. The data are discussed in terms of various theoretical models for the electronic structure of liquid semiconductors. It is shown that nuclear relaxation-rate measurements provide a means for characterization of the microscopic electronic transport mechanism. In the case of In_2Te_3 and Ga_2Te_3 an unusual enhancement of the nuclear relaxation rate relative to the predicted Korringa rate is shown to be consistent with the existence of localized electronic states at the Fermi level. Sb_2Te_3, on the other hand, exhibits no appreciable enhancement and is characterized as a metallic liquid. A general scheme is proposed for classification of the electron dynamics in electronically conducting liquids by the correlation of the nuclear relaxation rate and the dc conductivity.

I. INTRODUCTION

A number of electronically conducting liquids have been described as "liquid semiconductors" because their transport properties are reminiscent of those found for ordinary crystalline semiconductors. Although the properties of these liquids are widely varied, some general characteristics may be said to be typical of the group.[1] For example, most of the known liquid semiconductors are binary alloys or compounds consisting of a metallic component and a chalcogenide (O, S, Te, or Se) and they are invariably semiconducting in the solid phase. The liquids exhibit electrical conductivities σ less than about 10^3 $(\Omega\,\mathrm{cm})^{-1}$, and the conductivity increases with increasing temperature. In some cases, the temperature dependence of σ for the liquid is essentially a continuation of that of the corresponding solid, while in others (notably those with higher values of σ) the temperature variation changes markedly at the melting point T_m. The Hall coefficients have been measured for a few liquid semiconductors and have been found to be negative and significantly larger than free-electron values calculated for the total number of valence electrons. The Seebeck coefficients exhibit complex behavior in that they may be either positive or negative and, in fact, they often change sign with variations in temperature or alloy composition.

Theoretical understanding of the properties of liquid semiconductors in terms of their electronic structure is still at an early stage. The transport properties show clearly that these liquids are very different from ordinary liquid metals and metallic "molten semiconductors," such as Si or InSb. On the other hand, it is by no means evident that conventional semiconductor concepts derived for crystals may be extended to liquids without major modifications. This situation has led to new and, in some respects, conflicting speculations concerning the electronic structure of liquid and amorphous-solid semiconductors. The models differ particularly in their assumptions concerning the localized nature of the electronic eigenstates and details of electronic transport. These assumptions concern phenomena which are microscopic in that they occur over distances comparable to the interatomic spacing and, unfortunately, they are difficult to test directly by measurements of bulk transport properties at elevated temperatures.

In this paper we describe the application of nuclear magnetic resonance (NMR) to the study of liquid semiconductors. We show that the sensitivity of NMR experiments to details of the microscopic electronic and molecular dynamics yields important information not available from transport measurements. Specifically, we will report and discuss measurements of the Knight shifts and nuclear relaxation rates for In^{115} in liquid In_2Te_3, Ga^{69} and Ga^{71} in liquid Ga_2Te_3, and Sb^{121} and Sb^{123} in liquid