Neutron excitation of bulk luminescence

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The possibility of producing luminescence by using high-energy neutrons in a doped semiconductor is investigated. It is found that the analysis of the luminescence spectrum could allow for a measure of the spectral function of the excited electron within the bulk of the sample.

Recently it has been shown[1] that neutrons of relatively high energy can excite electrons in solids giving rise to a process similar to the more familiar photoemission, even though the neutron-electron interaction is weak[2] and the neutrons interact with the bulk of the sample. More interesting appears the analysis of the decaying of the electrons excited within the bulk, considering that their probability of leaving the sample with little energy loss is negligible while the most probable process following the excitation is likely to be an electron-electron or an electron-phonon scattering. However, if unoccupied states exist below the excited electron state also a radiative process can take place. This phenomenon excited through various energy sources is well known as luminescence. In general, the decay of the excited electron takes place through an impurity state or is assisted by a phonon in a nonconducting system.

When the primary excitation is produced by an impinging photon the luminescence process is critically dependent on the energy of the incoming photon. Indeed if the photon energy is comparable with the material gap, the penetration depth of the incoming radiation is very small and the process is confined to a portion of the sample close to the surface. On the other hand, when hard photons are employed, the excitation process, as well as the consequent decay, are not well controlled since the exciting energy is too large as compared with the band gap. Therefore, neutron excitation appears to be rather interesting because it provides bulk excitation together with a controlled excited state.

As already shown,[1] in consequence of the large mass ratio between neutron and electron, a neutron of appropriate energy, usually fairly large, can excite electrons within a well-defined and restricted energy range. Moreover, the time-of-flight technique can be employed to deduce the incoming neutron energy when the radiative decay takes place in a short time. Finally, the neutron-induced luminescence process, if effective, could be employed to detect high-energy neutrons though the efficiency of a luminescence-based detector is probably very low. Nevertheless, the possibility of detecting the neutron-induced luminescence is being experimentally examined.[3]

The interaction between an impinging neutron and an electron giving rise to a subsequent radiative decay can be described as a second-order process by using the following perturbing potential:

$$H=H_{e\text{-}n}+H_{e\text{-}r} , \tag{1}$$

where $H_{e\text{-}n}$ represents the electron-neutron interaction[2] already studied[1] and $H_{e\text{-}r}$ is the interaction of the system with the photon field. This process as usual takes place through an intermediate state, as no first-order transitions can be obtained by means of the interaction Hamiltonian of Eq. (1). The transition rate per unit energy is given by

$$\frac{dW}{d\varepsilon}=\frac{2\pi}{\hbar}\sum_F \rho_F(\varepsilon)|K_{F0}|^2\delta(E_F-E_0)\frac{j_0\Omega}{v_0} , \tag{2}$$

where j_0 and v_0 are, respectively, current density and velocity of the incoming neutrons and Ω is the system volume. E_0 and E_F are initial and final total energies of system, neutron, and radiation. $\rho_F(\varepsilon)$ is the density of final states and the sum runs over all the final states containing one photon of a given energy. K_{F0} is the transition matrix element from the initial state $|0\rangle$ to the final state $|F\rangle$ which is given by

$$K_{F0}=\sum_i \frac{\langle 0|H|i\rangle\langle i|H|F\rangle}{E_0-E_i} , \tag{3}$$

where $|i\rangle$ is an intermediate state with energy E_i. We note that, in view of the structure of H, the matrix elements of Eq. (3) are nonzero for the two intermediate states: (i) one neutron in the final state, no photon and the system in whatever state, (ii) one neutron in the initial state, one photon and the system in whatever state. Moreover, we observe that both the states having $E_i=E_0$ and $E_i\neq E_0$ will contribute to Eq. (3) and that the case $E_i=E_0$ should be treated with some care because of the divergence of K_{F0}. Such a case can be treated by splitting K_{F0} into two contributions:

$$K_{F0}=K_{F0}^{(\mathrm{nd})}+iK_{F0}^{(d)} , \tag{4}$$

where

$$K_{F0}^{(\mathrm{nd})}=\mathrm{P}\sum_i \frac{\langle 0|H|i\rangle\langle i|H|F\rangle}{E_0-E_i} , \tag{5a}$$

and

$$K_{F0}^{(d)}=\mathrm{Im}\sum_i \frac{\langle 0|H|i\rangle\langle i|H|F\rangle}{E_0-E_i-i\Gamma_i/2} . \tag{5b}$$

P indicates that the principal value has to be taken and Γ_i is the total inverse lifetime of the intermediate state. The energy diagonal term $K_{F0}^{(d)}$ in Eq. (4) is generally larger than the nondiagonal one $K_{F0}^{(\mathrm{nd})}$, therefore we shall confine ourselves to this term, usually named the *on-energy-shell* contribution. Since the $K_{F0}^{(d)}$ term is particularly large when $E_i=E_0$, Eq. (2) can be written as

$$\frac{dW}{d\varepsilon}=\frac{2\pi}{\hbar}\rho_F(\varepsilon)\delta(E_F-E_0) \times\frac{j_0\Omega}{v_0}\sum_i \frac{|\langle 0|H|i\rangle\langle i|H|F\rangle|^2}{[(E_0-E_i)^2+\Gamma_i^2/4]^2}\Gamma_i^2/4 \tag{6}$$

if the matrix elements do not strongly depend on the energy. Equation (6) contains the implicit factorization of the excitation and emission processes, an approximation employed in different cases.[4–6] As it has been observed in Refs. 4–6, such a factorization is a good approximation when the lifetime of the excited states is short as compared with the time of the emission, i.e., when the radiative-decay contribution to the width of the intermediate states is small.

The radiative transition rate per unit energy from state $|i\rangle$ to final state $|F\rangle$ is given by

$$\frac{dW_i^{\mathrm{rad}}}{d\varepsilon}=\frac{2\pi}{\hbar}\rho_F(E_F)\delta(E_F-E_i)|\langle i|H|F\rangle|^2 . \tag{7}$$

Moreover, the transition rate per unit energy in exciting the system from its ground state to the excited state $|i\rangle$ is[1]

$$\frac{dW_i^e}{d\varepsilon}=\frac{2\pi}{\hbar}\rho_e(E_0)\delta(E_i-E_0)\frac{j_0\Omega}{v_0}|\langle 0|H|i\rangle|^2 , \tag{8}$$

where $\rho_e(E_i)$ is the total density of states with energy E_i. The radiative contribution Γ_i^{rad} to the inverse lifetime of the state $|i\rangle$ is obtained by integrating Eq. (7) over the energy, so when using Eq. (8) the transition rate is given by

$$\frac{dW}{d\varepsilon}=\sum_i{}^{(d)}\Gamma_i^{\mathrm{rad}}\frac{dW_i^e}{d\varepsilon}\frac{1}{\pi\rho_e(E_0)}\frac{(\Gamma_i/2)^2}{[(E_0-E_i)^2+(\Gamma_i/2)^2]^2} . \tag{9}$$

This equation, which is accurate only when the on-energy-shell contribution is large, is particularly suitable when different excitation processes are considered as the use of the appropriate rate $dW_i^e/d\varepsilon$ is simply required. In Eq. (9) the sum runs over all the states on the energy shell, i.e., all the states having the same energy of the initial state. Therefore it can be performed by averaging the matrix elements of all the states having the energy within a range of the order of Γ_i around E_0, thus getting

$$\frac{dW}{d\varepsilon}=\left\langle\frac{\Gamma_i^{\mathrm{rad}}}{\Gamma_i}\frac{dW_i^e}{d\varepsilon}\right\rangle . \tag{10}$$

Equation (11) could have been directly obtained within a rather naive picture if one assumes that excitation and emission are completely decoupled processes. However, this approach is meaningful only if the excitation cloud around the hole present in the intermediate state can relax before the emission takes place.[7]

A direct insight into the kinematical constraints present in the neutron-induced luminescence process and a quantitative estimate of the transition rate can be obtained by resorting to some simple model for the system. The simplest approximation to the electronic structure of a doped semiconductor is given by that of a homogeneous noninteracting electron gas with an energy gap between occupied and empty states and the acceptor energy level close to the top of the valence band. In the present model we neglect the donor states to simplify the formal treatment. The use of incoming neutrons of finite wave vector $\mathbf{k}_0$ gives access to an immediate description of the indirect transitions without taking into account phonon-assisted transitions which are treated elsewhere.[8] Quantitative evaluation of the transition rate given by Eq. (10) for this simple model system follows directly by integrating the $dW_i^e/d\varepsilon$ term over the possible directions of the excited electron wave vector $\boldsymbol{\chi}$. The calculation of $dW_i^e/d\varepsilon$ was performed in Ref. 1 for the case of N-independent fermions under the assumptions of nonpolarized incoming neutrons and of cubic paramagnetic system and has been easily extended to the present semiconductor model. In such a case the average appearing in Eq. (10) can be done analytically, even though the explicit results are not presented here as they are very involved.

A further check on the validity limits of the present semiconductor model can be done by calculating the transition rate of Eq. (10) in the case of photon-induced luminescence which is a well-investigated phenomenon. The appropriate $dW_i^e/d\varepsilon$ term is thus obtained by using the interaction Hamiltonian

$$H_{e\text{-}r}=-\frac{e}{mc}\sum_i \mathbf{A}(\mathbf{r}_i)\cdot\mathbf{p}_i \tag{11}$$

in Eq. (8), $\mathbf{A}$ being the vector potential of the electromagnetic field and $\mathbf{p}$ the electron momentum. As it is known,[9] no electronic transition takes place in the case of an independent electron gas as the energy and momentum conservation cannot hold at the same time. However, the introduction of an energy gap in the electronic structure makes the process possible. Then, after some algebra, one gets

$$\frac{dW_i^e}{d\varepsilon}=\frac{\alpha j_0^{\mathrm{ph}}\Omega\chi^3}{2\pi\omega^{\mathrm{ph}}mc}\sin^2(\theta) \times\delta\left[\hbar ck_0^{\mathrm{ph}}+\left[\frac{\hbar k_0^{\mathrm{ph}}}{2m}\right]^2-\frac{\hbar^2\mathbf{k}_0^{\mathrm{ph}}}{m}\cdot\boldsymbol{\chi}\right] \times\Theta(k_f-|\boldsymbol{\chi}-\mathbf{k}_0^{\mathrm{ph}}|) ,$$

where j_0^{ph}, $\hbar\omega^{\mathrm{ph}}$, and $\mathbf{k}_0^{\mathrm{ph}}$ are, respectively, current densi-

ty, energy, and momentum of the incoming photon, α is the fine-structure constant, k_f is the Fermi momentum set at the top of the valence band, and θ is the angle between $\mathbf{k}_0^{\rm ph}$ and χ. As in the case of the neutron-induced process this quantity has to be integrated over the directions of the wave vector χ, which still can be done analytically. In any case, from the last equation it can be seen that an electronic transition takes place only if the photon energy is almost equal to the energy gap.

The determination of the luminescence rate implies an estimate of both radiative and total inverse lifetimes $\Gamma_i^{\rm rad}$ and Γ_i. To evaluate $\Gamma_i^{\rm rad}$ we used the standard formula for the radiative transition rate,[4] assuming a hydrogenic wave function for the impurity states. We get

$$\Gamma_i^{\rm rad}=\frac{4096\pi}{3}\alpha(E_{\rm ph})^3\frac{1}{(\hbar c)^2(\lambda^2+\chi^2)^6}N_I\chi^2\lambda^5\ , \qquad (12)$$

where λ is the decay constant of the hydrogenic wave function, $E_{\rm ph}$ is the energy of the emitted photon, and N_I is the density of impurity states. The evaluation of Γ_i is much more complex because various interactions contribute to the decay of electron states. To the purpose of the present estimate we can assume an energy independent Γ_i of the order of 10^{-3} eV.[10]

By using the above estimates we evaluated the luminescence rate by assuming the following parameters: $j_0=10^9$ neutrons/cm^2 s, $k_f=0.5$ a.u.$=0.945$ Å^{-1}, $N_I=10^{21}$ cm^{-3}, and $\lambda=1$ Å^{-1}. The results are reported in Fig. 1 at the two incoming neutron energies 300 and 500 eV by setting the energy of the acceptor level 0.1 eV above the highest occupied state. The photon-induced luminescence rate calculated by using the same estimate of $\Gamma_i^{\rm rad}$ and Γ_i and $\hbar\omega^{\rm ph}=1.001$ eV results in, as expected, a rate of several orders of magnitude greater than the neutron luminescence rate. It should be noted that the photon-induced luminescence is confined to a thin layer close to the sample surface, while our estimates assume no attenuation of the incoming photon beam. However, even if we take into account the lower depth relevant in photon excitation the neutron excitation is always much weaker.

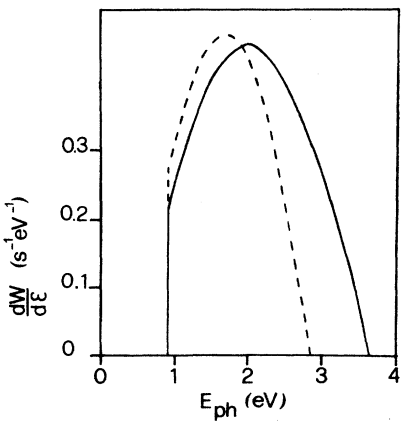

FIG. 1. Luminescence rate at 300 eV (dashed line) and 500 eV (solid line) incoming neutron energy.

We note that the radiative lifetime we obtain ranges from 10 to 100 ns, thus being negligible as compared with the neutron time of flight, which is 2.5 μs per meter of flight path when the energy is as high as 1000 eV. As a consequence the measurement of the photon spectrum versus time allows the determination of the energy of the incoming neutron. Since the photon energy defines also the excited electron energy, the acceptor level being known, the final neutron energy can be deduced. Therefore the neutron-induced luminescence is a measure of the spectral function of the excited electron integrated over the electron wave vector at constant energy. Then, analyzing the luminescence spectrum, it is possible to infer some information about the excited electron state *within the bulk*. Looking at Fig. 1 we see the real limitation of the neutron-induced luminescence, that is a quite low photon emission rate. Nevertheless, the information contained in the emission rate is rather appealing and we believe that an experimental study would be worthwhile.

Although the previous discussion opens up the possibility of real experiments, the analysis of background sources is worthwhile. Indeed the presence of stray γ radiation can seriously affect the possibility of the experiment even though a proper shielding as well as the analysis of the time-of-flight spectra could reduce the effect of *external* γ background within a negligible level. In addition, the presence inside the sample of isotopes having an appreciable cross section for n-γ reactions can preclude completely the experiment.

Apart from the above effect, another *intrinsic* spurious source is present. In fact, the strong neutron-nucleus interaction at neutron energies as high as 500 eV can produce excitation of the lattice. The effect of such an interaction is the creation of a photon cloud or the displacement of a nucleus, as in the present energy range the neutron can release to the lattice up to about 30 eV, 20 eV being the average released energy. Such a value has been deduced by assuming a nuclear mass 70 times that of the neutron in the hypothesis of free-particle collision. In a semiconductor the phonon cloud will relax mainly through the interaction with the lattice, while the direct photon-electron interaction is essentially negligible. On the other hand, if a nucleus is displaced as a consequence of the collision it can be considered as a charge of non-negligible energy moving through the lattice and bringing a strong electromagnetic interaction so that particle-hole pairs can be created. Nevertheless, the same kinematical constraints which make the total electromagnetic neutron cross section very small, also make the cross section of a slowly moving heavy charge very small. We first observe that the total electromagnetic neutron cross section,[2] neglecting kinematical limitations, is of the same order of the nuclear one, i.e., $4\pi(\gamma r_0^e)^2$, r_0^e being the electron classical radius. Therefore the ratio of the rates of electromagnetic and nuclear processes is proportional to the solid angle within which the neutron-electron interaction is allowed, whereas almost no limitation is present for the neutron-nucleus interaction. This solid angle turns out to be of the order of m/M, M being the neutron mass. On the other hand the interaction between an electron and a charged particle whose mass is M_n is confined

to a solid angle of the order of m/M_n, so that the nucleus-mediated luminescence processes are expected to be M/M_n times smaller than the direct ones.

As a consequence of the above discussion, we can assume that almost all the energy released by the neutrons through the nuclear interaction produces a lattice excitation which relaxes by interacting with the lattice itself. Such a mechanism will rise the local temperature thus increasing the number of thermally excited electrons. However, a reasonable estimate by using a phonon free path of 20 Å gives a local increase of 10^{-4} K over a fraction 10^{-5} of the total volume. Thus, very little increase of the photon emission in the eV range is expected to take place through the phonon excitation.

Finally we observe that, in view of the very low-emission rate, there is very little chance of using the present process to detect high-energy neutrons.

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