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DETECTION OF IMPURITY TUNNELING IN SOLIDS VIA COHERENT PHONON COUPLING AND DIRECT NEUTRON SCATTERING

R.C. Casella

National Measurement Laboratory, National Bureau of Standards, Washington, D.C. 20234, U.S.A.

Abstract.— A theoretical treatment is given of the observation of molecular tunneling in solids by the coherent interaction of the tunnel-split excitations with acoustic phonons and by direct neutron inelastic scattering. Results are applied to the case of rotation tunneling of CN⁻ dumbbells in KBr and KCl, and to the motion of H atoms in two-well traps associated with oxygen impurities in niobium. Comparison is made with experiment.

Coherent forward resonant scattering of phonons from the tunnel-split ground and librionically excited configurations of impurity molecules in solids results in mixed modes, with non-zero component amplitudes for both the phonon and the coherently excited impurity complexes. When observed via inelastic neutron scattering, one sees a perturbed acoustic branch in the neighborhood of the wave vector \vec{k} at which the component modes are degenerate [1], [2]. The mixed mode $|\Phi\rangle$ is given by relation (1), where $|EX, \vec{R}\rangle$ represents the impurity excitation at site \vec{R} .

$$|\Phi\rangle = |\text{phonon}, \vec{k}\rangle + i \sum_{\vec{R}} |EX, \vec{R}\rangle \langle EX, \vec{R}|T|\text{phonon}, \vec{k}\rangle \tag{1}$$

where the sum on \vec{R} of the T-matrix elements extends only over lattice sites occupied by the dumbbell impurities. The mixed modes have been described in terms of vector cubic harmonics and analyzed group theoretically in terms of a model [2]

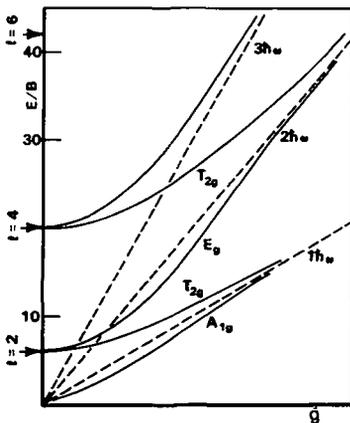


Fig. 1. Energy level schematic for the unperturbed impurity complex: the A_{1g} lowest level and the neutron-active low-lying E_g and T_{2g} states vs. the coupling strength g of the hindering potential for the model in which the impurity dumbbells are aligned along $\langle 111 \rangle$ directions. The first excited librionic level is of type E_g . The tunnel split excitation within the librionic ground state is of type T_{2g} . B is the rotation constant; l is the angular-momentum quantum number when $g = 0$.

which accommodates the neutron scattering data [3], [4] as well as the infrared and Raman scattering data [5] for KCl:CN^- and KBr:CN^- . The more easily observed phonon admixture with the first vibronically excited tunnel state is determined to be of E_g character. (See Fig. 1.) If the tunnel-split second excited state (T_{2g} character) were to lie close to the E_g excitation in these systems it ought couple with equal strength [2]. The inability to observe the T_{2g} interaction via neutron scattering indicates that it lies above the potential barrier and is subject to lifetime broadening due to multi-phonon emission. This destroys the coherence required for its observation via the neutron probe. The observation of a broad T_{2g} state in the Raman spectra considerably above the E_g [5] is in accord with this picture. Moreover, the model leads to the conclusion that the tunnel-split ground state in these systems ought be observable by neutron scattering (via its coupling to the acoustic phonon) only in the T_{2g} configuration (Fig. 1). This prediction has been confirmed experimentally in KBr:CN^- [3].

At low temperatures, dilute ($< 0(1\%)$ concentration) hydrogen atoms in metals such as niobium and tantalum, also doped with $0(1\%)$ concentration oxygen atoms, become trapped at the latter sites in what are believed to be two-well centers. Tunneling of the hydrogen between the two potential minima near the oxygen provides another interesting system in which molecular (atomic) tunneling in solids can be studied under quite different conditions. Because of the very large low-energy n p scattering length it is possible to study the excitations directly via neutron scattering. This has been done both for transitions among the tunnel-split components of the vibronic ground state [6] and also for transitions from them to the first and second vibronically excited states [7]. Here the picture is complicated by local strains which can produce relative displacements in the energy of the component well minima. This greatly reduces the ratio of inelastic to elastic neutron scattering within the ground doublet according to a model developed in [6]. The reduction is by a factor $\frac{1}{\sin^2 2\theta}$ where θ is a mixing angle between the left and right single-well states, describing their admixture in tight-binding approximate tunneling eigenstates. (The average is over some assumed distribution of the strain induced relative displacements.) In principle, for zero strain (infinite dilution) the elastic and inelastic differential cross sections for scattering within the ground doublet ought to be comparable. It is important to understand theoretically the neutron induced transitions to the tunnel-split excited states in order to confirm the model directly. If separated considerably from the ground doublet, the excited doublet can be characterized by a mixing angle ϕ , generally different from θ . Matrix elements for transitions from either component of the ground doublet to either of the tunnel-split excitations depend upon both θ and ϕ . Under suitable experimental conditions, however, the differential cross section for excitations from both components of the ground doublet to either component of the excited doublet can be shown to be independent of θ and of ϕ . Thus the ratio Λ of inelastic cross sections,

$$\Lambda = \frac{d\sigma/d\Omega^{\text{excited}}}{d\sigma/d\Omega^{\text{ground}}} \frac{\text{inelastic}}{\text{inelastic}}, \quad (2)$$

is enhanced by a (concentration dependent) factor $1/(\sin^2 2\theta)$. The numerator in the expression for Λ is diminished by the transition form factor $\tilde{F}(\vec{q}')$ where \vec{q}' is the momentum transfer in the higher energy inelastic neutron scattering experiment involving vibronic excitation. For small \vec{q}' , $\tilde{F}(\vec{q}') \sim (\vec{q}' \cdot \vec{D})^2$, where \vec{D} is the dipole transition matrix element. For small momentum transfer \vec{q} in the lower energy experiment involving transitions within the ground doublet, the denominator $\sim (\vec{q} \cdot \vec{r}/2)^2$ where \vec{r} denotes the spatial displacement of the well minima within each two-well complex. Thus, in the limit $\vec{q}' \rightarrow 0$, $\vec{q} \rightarrow 0$ such that ratio ρ of the larger to the smaller remains fixed, Λ remains finite. I find, in this limit,

$$\Lambda = (\rho^2/8) (k_F/k_I) (\hbar\omega/V_0) / (\sin^2 2\theta). \quad (3)$$

Here, (k_F/k_I) is the ratio of the final to the initial momenta of the neutron in the higher energy experiment, $\hbar\omega$ is the vibronic excitation energy, and V_0 is the barrier height between the potential minima. General expressions have also been derived for Λ and various other cross section ratios when the small \vec{q} , \vec{q}' expansion is not justified [8]. Because of the disparate conditions under which the higher [0(100 meV)] and lower [0(0.2 meV)] energy neutron scattering experiments are run in Nb:0(1%) H, 0(1%) O, many difficult normalization problems have to be dealt with in order to determine Λ even semiquantitatively [7]. Nevertheless, it would appear worthwhile.

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