

ENHANCEMENT OF FUSION RATE INDUCED BY THE COLLECTIVE ELECTRON EXCITATIONS

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After the claimed experimental evidence of deuteron cold fusion reported in the literature¹, a large amount of theoretical work has been done, either to support the possibility of this unexpected phenomenon, either to present arguments against it. The paper by Legget and Baym² appears of fundamental relevance to this respect. In that work a rigorous upper bound is given for the fusion rate of deuterons embedded in a crystal, which is too small to be compatible with the claimed experimental values. The upper bound is expressed mainly in terms of the chemical potential of neutral helium atoms and deuteron molecules with respect to a crystal of Palladium, or in general of the absorbing material. The smallness of this upper bound is a consequence of the small value of the chemical potential of neutral helium atoms. The latter has been taken to be close to zero, according to the experimental evidence. Thus the result appears unavoidable, and in agreement with this finding some authors have suggested unconventional phenomena to give support to the anomalous fusion rate. However, within the conventional solid state theory, there is still a possibility. One can in fact postulate that the interaction energy between helium atoms and the Pd crystal possesses a second minimum, separated from the till now observed one by a large potential barrier, and at lower energy. This minimum could be practically unreachable by the usual experiments with neutral helium atoms interacting with the crystal, because of the large potential barrier, but it could be easily reached along the rather different path followed by the deuteron–deuteron fusion process. In this contribution we explore this possibility in detail, by studying the interaction of a positive charge with the electrons of a crystal of Pd. The response function of an electron gas can be described in terms of its possible excitations. A positive charge of mass comparable with the mass of the deuteron can be considered as a fixed charge, since its recoil energy is negligible with respect to the energy of the electron excitations. Let us consider the coupling of the charge with one of the excitation mode. We schematize the latter with a simple bosonic branch of energy $\hbar\omega(q)$, with $q \leq q_c$. The cutoff q_c is of the order of $2\pi/a$, a being the average lattice spacing. Then the system is described by the hamiltonian

$$\mathcal{H} = \sum_q \hbar\omega(q) a_q^\dagger a_q + \sum_q v(q)(a_q^\dagger + a_q) \quad (1)$$

The *exact* ground state of the hamiltonian (1) is the product of coherent states $|\alpha_q\rangle$, one for each mode q , with energy E

$$|\Psi_0\rangle = \prod_q |\alpha_q\rangle \quad ; \quad \alpha_q = -v(q)/\hbar\omega(q) \quad (2)$$

$$E = - \sum_{q \leq q_c} \frac{v(q)^2}{\hbar\omega(q)} \quad (3)$$

The same result can be obtained by using perturbation theory for the charge particle energy, in the limit of large mass. The first non-vanishing contribution is the *second order* term, which gives $E = - \sum_{q \leq q_c} v(q)^2/q^2/2M + \hbar\omega(q)$. For $M \rightarrow \infty$ one regets the result of eq. (3). This also shows that the perturbation theory actually has zero radius of convergence, as one can verify directly by considering higher order terms of the energy expansion. It has to be noticed, however, that the second order wave function can be quite different from the exact one. A third way of getting the result of eq. (3) for the ground state energy is to use the expansion of the self-energy of a particle in the framework of the dielectric formulation of many-body theory. The dielectric function $\epsilon(q, \omega)$ of the system of bosons can be written

$$\epsilon^{-1}(q, \omega) = \frac{\omega^2(q)}{\omega^2 - \omega^2(q)} \quad (4)$$

The self-energy $\Sigma(q, \omega)$ of a particle moving in the system can be expanded in powers of the dielectric function. Again, to lowest order, one regets eq. (3), provided one regards $\Sigma(q, \omega)$ as the energy of the particle and take the limit $M \rightarrow \infty$ and consistently the unperturbed particle energy $E = 0$ (Raleigh-Schrödinger perturbation theory). This last formulation of the problem is quite useful, because it can be generalized to more realistic case, where the collective excitations are not schematized by simple bosons, but properly described within the many-body theory of the electron system. In this case the dielectric function of eq. (4) is substituted by more microscopic expressions. In view of the above discussion, however, it is likely that also in this microscopic formulation the perturbation expansion is divergent, and it is apparent that the best strategy to adopt is to calculate, also in this case, the energy to lowest order, since it should give a result rather close to the exact one. The main feature of the result of eq. (3) is the dependence of the energy on the square of the coupling $v(q)$. It follows that a particle of charge Q has an energy E proportional to Q^2 , $E = \beta Q^2$, $\beta < 0$. Consequently two separated particles, each one of charge Q , have a total interaction energy $E_{1+1} = 2\beta Q^2$, which is a factor two smaller in absolute value with respect to the case they are close to each other to form essentially a single particle of charge $2Q$, $E_2 = 4\beta Q^2$. The decrease in energy is $E_2 - E_{1+1} = 2\beta Q^2$. Thus the coupling with the electron collective excitations produces an attractive potential between two charged particles. This is well known in the theory of charge screening in solids³, but it has to be noticed that it is a more general phenomenon. For instance the phonon exchange, which produces the Cooper pairs in ordinary superconductors,

has a similar origin. For an electron system the dielectric function can be parametrized as

$$\epsilon^{-1}(q, \omega) = \sum_i \frac{S_i}{\omega^2 - \omega_i^2(q)} \quad (5)$$

where the strengths $\{S_i\}$, one for each mode i , are constrained to fulfil the f -sum rule (or "oscillator sum rule")

$$\int_0^{+\infty} d\omega \text{Im}(\epsilon^{-1}(q, \omega)) \omega = \frac{\pi}{2} \sum_i S_i = \frac{2\pi^2 e^2 n}{m} \quad (6)$$

where n is the electron density and m the electron mass. For a purely collective excitation $S_i = \omega_i^2$. In general one can assume $S_i = s_i \omega_i^2$, with $0 \leq s_i \leq 1$, and the sum rule reads

$$\sum_i s_i \omega_i^2 = \frac{4\pi e^2 n}{m} = \omega_p^2 \quad (7)$$

Once the frequencies $\{\omega_i\}$ are given, this relationship constrains the possible values of the weights $\{s_i\}$, as it will be discussed later on. In general the momentum q of each excitation has a cut-off value, above which the excitation becomes overdamped. For simplicity one can assume the same cut-off q_c for all the excitations. According to eq. (3) the energy of a particle of charge Ze is given by

$$E_Z = -\frac{(Ze)^2}{\pi} q_c \left(\sum_i s_i \right) \quad (8)$$

and the corresponding gain in energy ΔE for "fusion" of two particles, each of charge Z , is twice this value (for $d-d$ fusion $Z=1$). From the constraint (7) one gets

$$\sum_i s_i = \omega_p^2 / \bar{\omega}^2 \quad ; \quad \bar{\omega}^2 = \frac{\sum_i s_i \omega_i^2}{\sum_i s_i} \quad (9)$$

It follows that the total strength $\sum_i s_i$ is larger if the average energy $\bar{\omega}$ of the electron excitations is smaller. This result has a simple physical interpretation: a smaller $\bar{\omega}$ implies a larger polarizability and thus a stronger interaction energy of a charged particle with the medium. If one assumes a dispersionless frequency ω_i , it is possible to get an analytical expression for the potential energy between two particles of charge Ze at a distance R , due to their coupling with the electron system. Following ref.⁴ one gets

$$V(R) = -2 \frac{(Ze)^2}{\pi} q_c \left(\sum_i s_i \right) I(q_c R) \quad ; \quad I(x) = \frac{1}{x} \int_0^x \frac{\sin(y)}{y} dy \quad (10)$$

Before discussing the implications of this result for the specific case of Pd, let us consider some possible criticisms to the validity of eqs. (10). First of all the dielectric function of eq. (5) does not display the zero at $\omega = 0$, $q \rightarrow 0$, which characterizes the dielectric function of a metal, the so called "complete screening". This is due to

the fact that eq. (5) does not include the single particle conduction band transitions, which are responsible for the static screening in a metal. Their inclusion should affect only a range of momenta around $q = 0$ of the static inverse dielectric function $\epsilon^{-1}(q, 0)$ and the result (8) should be essentially unaffected. Another warning about the dielectric function of eq. (5) comes from the observation that, as a function of q and for $\omega = 0$, it is negative in the relevant momentum range. This fact could cast some doubts on its validity, since it could be in contrast with the stability condition of the crystal. For a deformable jellium model, in fact, negative values of $\epsilon^{-1}(q, 0)$ indicate that the system is unstable to density fluctuations⁵, which are not observed in transition metals. It has to be kept in mind however that the dominant contribution to the crystal stability is due to the Ewald energy⁶, which originates from the non-jellium structure of the crystal. Eq. (5) has to be considered as an average of the dielectric function on orientation of the wave vector \vec{q} . The instability coming from the negative values of $\epsilon^{-1}(q, 0)$ can be counter-balanced by the stabilization due to the Ewald static energy from the crystal structure. The experimental data on inelastic electron scattering on Pd of ref.⁷ are reported in fig. 1, where the second derivative spectrum is shown as a function of the excitation energy.

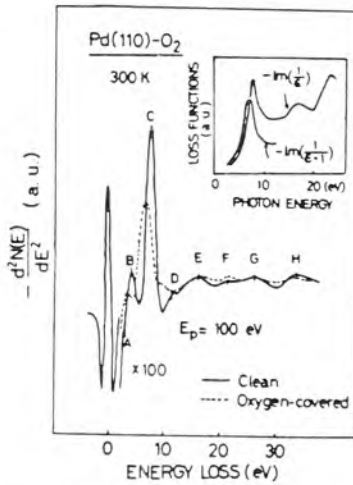


Fig. 1 Electron energy loss spectrum (full line) from ref. 7.

Several peaks are observed. Even if some of them could be simple atomic transitions, nevertheless they are associated with transitions in the sd valence band. According to ref.7 the two peaks at $\omega_1 = 7.5$ eV and $\omega_2 = 26.5$ eV correspond to plasmon excitations mainly of the s and d band respectively. Accordingly the broad peak observed at $\omega = 33.8$ eV, quite close to $\omega_1 + \omega_2$, must be interpreted as due to the simultaneous (second order) electron excitation of both the s and d plasmons. Therefore the bump does not correspond to an independent excitation, and its energy has not to be included in the sum of eq. (7). The theoretical justification of the two independent plasma excitations of the s and d bands can be the smallness of the particle-hole matrix elements between the two bands, which involve different orbitals with a small overlap. Using the Pd electron density $n \approx 0.4 \text{ \AA}^{-3}$, one gets $\sum_i s_i \omega_i^2 \approx 10^3 \text{ eV}^2$, and, according to the preceding discussion, eqs. (7) gives $\sum_i s_i \approx 6$. The corresponding $d-d$ potential of eqs. (10) is depicted in fig. 2. The value of $q_c = 2.2 \text{ \AA}^{-1}$ has been assumed. In ref.⁸ the branch at $\omega = 7.5 \text{ eV}$ has been observed up to $q \approx 1 \text{ \AA}^{-1}$ with essentially no

dispersion. No data on the other excitations are available in the literature.

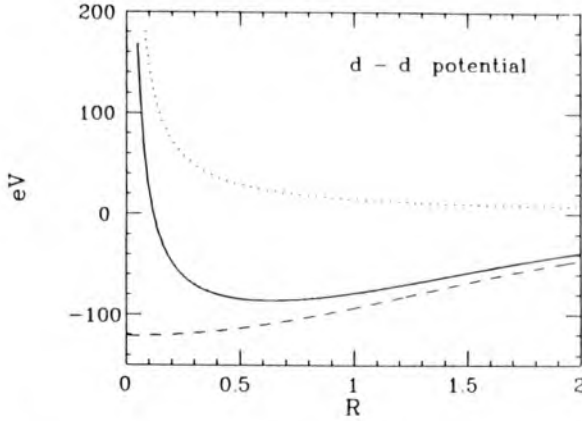


Fig. 2 Interaction potential between two deuterons (full line). The dotted and dashed lines are the coulomb and electron contribution respectively. The distance R is in \AA .

The large interaction energy is a consequence of the large charge electron displacement around the positive charge. The latter can be calculated from the dielectric function of eq. (5) and is reported in fig. 3 as a function of the distance from the positive charge. The total polarization charge concentrated in the first large peak depends only on the strength $\sum_i s_i$, and it exceeds largely the positive charge in this case. In that sense one can speak of "overscreening" of the positive charge. The phenomenon is possible due to the collectivity of the excitations, which prevents the electron kinetic energy to increase too much as the electron charge is localized, since the collective excitations have a large "mass". It must be stressed that the above discussed correction of the dielectric function near $q = 0$ modifies the large distance behaviour of the polarization charge, and it is necessary in order to ensure that total displaced charge, obtained integrating up to infinity, is equal to the positive charge, according to a general property of the electron gas⁹. The fusion rate R can be calculated by considering a deuteron gas interacting through the potential of eqs. (10), following the same line of ref. 3. The result gives

$$R \approx 1 \text{ fusion/sec./mole} \quad (11)$$

The rate R is of course sensitive to q_c . The assumed value $q_c = 2.2 \text{\AA}^{-1}$ appears realistic. The "delocalized" configuration described here of a positive charge interacting with the electron collective excitations is quite different from the one corresponding to an atom-like configuration where the positive charge is surrounded by its electrons, essentially as in its free configuration. For helium the delocalized configuration is largely favourable in energy with respect to the atom-like one. However an helium atom inside the crystal cannot decay spontaneously to the delocalized configuration. Suppose in fact to polarize the helium atom to bring it to the delocalized configuration. On one hand this requires some energy, and on the other hand the interaction, with the electron gas, of the so formed dipole produces a gain in energy, according to the above described model. Which one of the two energy changes actually dominates is determined by the curvature of the energy surface. The latter can be estimated as E_B/a^2 , where E_B is the binding energy and a the size of the system. It turns out that for helium atom the curvature is much larger, because of its small size, despite the

smaller binding energy. This is the physical origin of the potential barrier between the two configurations.

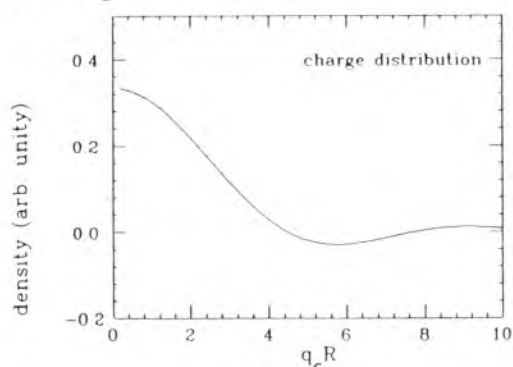


Fig. 3 *Electron charge displacement around a positive charge .*

The same reasoning can of course be followed for the deuteron (or hydrogen) atoms. In this case however the size of the atom is larger, and the two curvatures may be comparable. Thus, according to the model, the second minimum might be reachable under favourable conditions. The experimental observation of this second minimum for deuteron (or hydrogen) atoms would strongly support the model. In conclusion the model presented here explains a fusion rate of the order given by eq.(11). Furthermore it predicts the formation of deuteron-like molecules inside the crystal, sitting at the minimum of the potential of fig. 2. This implies additional heat production (for both hydrogen and deuteron atoms), whose rate, however, is determined by the approach to equilibrium (towards the bottom of the potential). A first attempt to describe the dynamics towards equilibrium was presented in ref.¹⁰, but it requires further investigations.

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