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FUZZY LOGIC IMPLEMENTED AT THE MOLECULAR LEVEL

Future Information Technology Systems will hinge on logic gates implemented at the molecular level. To expand the intelligence quotient of next artificial machines, it is necessary to find out how to process Fuzzy logic at the molecular level. Fuzzy logic allows certain and uncertain information, objective and subjective knowledge to be dealt with.

owadays, among the main issues that mankind has to face up there are those of Energy and Computation. It is compelling (1) to find out effective ways of exploiting renewable energy resources, such as solar energy, to cope with evergrowing clean energy demand and (2) to improve the computation capabilities of our artificial machines. Regarding this second goal, it is evident that there is a worldwide effort to design and improve devices and machines able to store, process and convey information more and more effectively. Current Information Technology Systems revolve around logic gates sculpted from bulk semiconductors that employ electronic input and output signals. The performances of the electronic circuits can be improved by shrinking their components and cramming logic gates onto smaller and smaller wafers of silicon. Their miniaturization is currently pursued by the top-down approach through photolithography and related techniques. However, when the bulk

semiconductor dimensions are reduced to nanometric size, fundamental scientific problems arise, such as current leakage and heat dissipation. Therefore, an alternative strategy, named as bottom-up approach, has been put forward over the last few years [1]. Its ultimate goal is to build a computer, similar in its basic operations to current silicon-based machines, with its underlying hardware based on single molecules, self-assembled supramolecular entities and/or chemical reaction networks. The development of molecular logic gates will allow not only electrical but also optical and other physical and chemical inputs and outputs to be exploited.

If the next molecular computers want to fulfil every need of computing, it is important to find out how to implement not only Boolean logic gates but also Fuzzy logic engines. Boolean binary logic has the peculiarity of manipulating only statements that are true or false, reducible to strings of zeros and ones. However, quite often, the available data and knowledge suffer a certain degree of uncertainty and imprecision, especially when they are based on subjective linguistic statements. In all these cases, it is still possible to process information by abandoning hard computing, based on binary logic and crisp systems, and adopting soft computing, based on Fuzzy logic, neural nets and probabilistic reasoning [2]. Fuzzy logic is likely to play an increasingly important role in the conception and design of systems whose machine intelligence quotient is much higher than that of systems designed by conventional methods, since it affords to deal with certain and uncertain information, objective and subjective knowledge. The most effective implementations of Fuzzy logic in electronics have been achieved by the use of analog electronic circuits that are based on continuously variable signals. In this paper we put forward ways of implementing Fuzzy logic at the molecular level after giving a glance at the ways of computing with molecules.

Molecular computing

In principle, it is possible to synthesize individual molecules that can act as diodes, transistors and other building blocks of microcircuits, based on electronic input and output signals. Such molecular electronic components would be about one million times smaller in area than the corresponding logic elements fabricated on a semiconductor chip. If this technology will one day succeed, ultrasmall and ultradense electronic computer will be built [1]. However, there are still some burdens to be overcome, such as the way of manipulating and ordering molecules in circuit-like structures and interfacing them with the macroscopic world.

Working with molecules allows not only electrical, but also chemical, optical and other physical inputs and outputs, such as heat and pressure, to be exploited for processing information. The ability of making "computation" by molecules resides in their structures and their reactivity (i.e. affinity). The order, the way the atoms of a molecule are linked, and their spatial distribution rule the intra- and intermolecular interaction capabilities of the molecule itself, defining its potentiality of storing, processing and conveying information. Molecules can work in a test tube, i.e. within a thermodynamically closed system, or in open systems, like the cells, exchanging chemicals with the environment. Closed systems make computations if they are perturbed from their equilibrium state. Molecules float around chaotically whereby it is a tough task to connect different molecular computational elements. If optical signals are involved in the computation, they do not readily allow one molecule to communicate specifically with another, since they are multidirectional. However, optical outputs have the great advantage of bridging the gap between the microscopic molecular world and our macroscopic world. Numerous molecular switches have been discovered so far whereby hard computing, based on photo/chemical codes, can be implemented. A survey can be found in some recent reviews [3]. All these switches process crisp Boolean logic by encoding information in the form of binary digits. For this purpose, it is necessary to establish a threshold value and a logic convention for every input and output variable. The variables can assume simply high or low values that become digital 1 or 0, respectively, in the positive logic convention, whereas the negative logic convention reverses this relationship. Bits are manipulated by logic gates, which transform crisp values in designed ways. The nature of logic gates that can be implemented depends on the response of chemical compounds to the physical or chemical inputs.

In order to rely on spontaneous appearance of macroscopically ordered structures, playing the role of communication channels between the microscopic and macroscopic worlds, molecules have to act in open systems. Open systems can evolve towards far-fromequilibrium steady states exhibiting long-range coherence. Spatiotemporal organizations, called "dissipative structures" by Prigogine [4], may emerge. They correspond to a form of supramolecular coherence involving an immense number of molecules, in contrast with equilibrium situations where the range of correlations is determined by short-range intermolecular forces. Every cell, that is the prototype of a complex far-from-equilibrium system, is a powerful molecular computer [5]. Its computations are based on cell signalling, that is the complex system of storing, processing and conveying information that governs basic cellular activities and coordinates cell actions. Enzymes play key roles in cell signalling, due to their power of turning on/off chemical reactions. Each protein recognizes a specific type of molecule and then switches its state by making or breaking a precisely selected chemical bond, like a key for its lock [6]. The fitting of key-enzyme to lock-substrate is a sophisticated pattern recognition operation based on the three-dimensional structures of the proteins and substrates.

Even DNA can compute. Its computing power derives from the ability of nucleotides to bind together using Watson-Crick pairing. There is a code of "stick" or "don't stick", with DNA chains either binding to form regions of double-stranded molecules or remaining free as regions of single-stranded DNA [7]. As Adleman pointed out [8], DNA hybridisation offers enormous parallel processing. While conventional computers attack problems via large calculations in series, properly encoded molecular computing systems might quickly solve the same problems by simultaneously carrying out billions of operations in parallel. The widest parallelism in molecular computing can be achieved by contriving devices operating in quantum mechanical regime. In the quantum world, any entity, that can exist under two distinct states, is a suitable candidate for implementing quantum computing: the up or down spin of an electron or a nucleus, the polarization of a photon [9], etc. The two possible configurations can correspond to the 0 or 1 of a bit. What makes the quantum bit, usually called qubit, peculiar is the possibility of setting it up as superposition of the 0 and 1 states at the same time. In other words, a gubit can be represented as a linear combination of $|0\rangle$ and $|1\rangle$:

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|\Psi\rangle = a|0\rangle + b|1\rangle
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Fig. 1 - The different ways of computing by molecules

The qubit "collapses" into a definite value as soon as it is measured. The probability of obtaining $|0\rangle$ is $|a|^2$, whereas that of collecting $|1\rangle$ is $|b|^2$. The superposition of states offers an enormous gain in the use of computational resources, since in general n qubits can store 2^n numbers at once. The superposition can involve also the quantum states of physically separated particles if they are entangled. The measurement of the state of one particle causes the collapse of its superposition of values. But the entangled particle then also instantaneously assumes the same value, no matter far apart the particle might be. There are a number of practical problems in building a quantum computer, deriving from the fact that quantum states must constantly contend with insidious interactions with the environment triggering loss of

coherence. To solve them, it is necessary to build a system, which is isolated from everything except the measurement and manipulation mechanisms (Fig. 1).

The principles of Fuzzy logic

Boolean logic allows only certain and objective information, based on numerical data, to be dealt with. To enlarge the computation capabilities of artificial machines, it is compelling to simulate human reasoning that is based mainly on vague linguistic statements. In 1965 Lofti Zadeh [10] proposed Fuzzy logic to handle both objective and subjective knowledge. Fuzzy logic allows the modes of the human reasoning and making decision in an environment of uncertainty and imprecision, to be modelled. At the hearth of the difference between classical and Fuzzy logic there is the law of excluded middle. In classical logic based on standard set theory, an object either does or does not belong to a set, fulfilling the principle of not contradiction. Fuzzy logic, based on Fuzzy sets theory, breaks the law of excluded middle, to some degree. Items belong only partially to a Fuzzy set and their degrees of membership can assume values included between 0 (absence of membership) and 1 (complete membership). They may even belong to more than one set. The only constraint on fuzzy logic is that, for an element, the degrees of membership in complementary sets must sum to unity. The law of the excluded middle holds merely as a special case in Fuzzy logic, namely when an element belongs 100 percent to one group. Fuzzy degrees are different from probability percentages. Probabilities measure whether something will occur or not. Fuzziness measures the degree to which something occurs or some conditions exist.

The entire domain of each variable, referred to as the universe of discourse, is divided into different Fuzzy sets whose shape and position define their membership

functions. For example, if the variable is the volume of water contained into a glass of 50 mL, its universe of discourse is defined by all water volumes from 0 to 50 mL and the word "full" would correspond to a curve that defines the degree to which any glass is full. If the set of "full glass" is given the well-defined boundary of a crisp set, we might state all glasses containing more than 40 mL are considered full. Such rigid boundary brings about unreasonable distinction between two glasses having 40.1 mL and 39.9 mL of water as full and not-full ones, respectively. The right way to define the set of full glasses consists in choosing a smoothly varying curve, the membership function, that passes from not-full to full. Every glass of water will belong to the "full glass" Fuzzy set in a certain degree (μ) included between 0 and 1. For instance, a glass with 42.5 mL of water



will belong to the "full glass" Fuzzy set with a degree of membership μ =0.9, whereas a glass containing 37 mL of water will have a μ (full glass)=0.5, according to the membership function depicted in Fig. 2. Of course, the subjective degree of thirst will influence the shape and position of "full glass" Fuzzy set along the universe of discourse of water's content inside the glass.

In order to process Fuzzy logic, a Fuzzy Logic System (FLS) must be implemented. A FLS is a non-linear mapping of an input crisp data vector (\bar{x}) into a scalar crisp output (y) and this mapping can be expressed quantitatively as $y = f(\bar{x})$ [11]. A FLS consists of three main components: a Fuzzy linference Engine and a Defuzzifier (see Fig. 3).

The hinge of a FLS is the Inference Engine that is based on Fuzzy rules, i.e. linguistic statements of the type "IF...THEN..." wherein the IF-portion, called the antecedent, involves the Fuzzy sets of the input variables, whereas the THEN-part, named as the consequence, involves the Fuzzy sets of the output variable. In case of multiple-antecedent, the different inputs can be connected through the AND, OR and NOT operators. The Fuzzifier maps crisp numbers into specific fuzzy sets of the input variables. It is needed in order to activate rules. The Inference Engine handles the way in which rules are combined. Just as we humans use many different types of inferential procedures to help us understanding things or to make decisions, so there are different Fuzzy logic inferential procedures. The Defuzzifier maps output Fuzzy sets into crisp numbers.

The Fuzzy Logic Systems can approximate any continuous mathematical function of cause and effect [12]; therefore, they are playing key roles in the development of machine intelligent quotient. They have been finding applications in numerous fields, such as in aircraft and cruise control (Rockwell Corp., Nissan), self-parking model car (Tokyo Tech. Univ.), space shuttle docking (NASA), elevator scheduling (Hitachi), stock market analysis, analytical chemistry [13], picture adjustment (Sony), handwriting recognition, video camera autofocus (Canon), to cite a few. The next challenge entails contriving strategies to implement Fuzzy Inference Engines at the molecular level in order to boost the sector of Molecular Computing.

Molecular Fuzzy Inference Engines

In the macroscopic world, the most effective implementations of Fuzzy logic in electronics have been achieved by the use of analog electronic circuits that are based on continuously variable signals. Since the microscopic world is quantized, at first sight it seems improbable that Fuzzy Logic Systems can be implemented at the molecular level. In the quantum world, only discrete states are available to particles. However, when two or more states are energetically and symmetrically accessible to a particle, the fuzzyness of quantum mechanics comes out. For instance, if a molecule can exist as a superposition of two states, represented by the wavefunctions ψ_1 and ψ_2 , its total wavefunction will be:

$$\psi_{\text{TOT}} = a |\psi_1\rangle + b |\psi_2\rangle$$

CRISP INPUTS

FUZZIFIER

FUZZY

INFERENCE

ENGINE

DEFUZZIFIER

and ψ_2 will act as Fuzzy sets of a variable representing the electronic states; the values a^2 and b^2 will represent the degrees of membership of ψ_{TOT} to the two Fuzzy sets.

Aromatic carbonyl and nitrogen-heterocyclic compounds are organic molecules exhibiting a quantum phenomenon whereby Fuzzy Logic Systems can be implemented. When these compounds absorb UV or visible photons of suitable frequency, they pass to an excited state that is a superposition of two wavefunctions, as indicated in equation (3). The two wavefunctions have π , π^* and n, π^* nature respectively, and involve primarily the C=O or the C=N group.

$$\psi_{\text{TOT}} = a |\psi_{\pi,\pi^*}\rangle + b |\psi_{n,\pi^*}\rangle$$

The two wavefunctions can vibronically couple. The extent of their coupling depends on the energy gap between them: the smaller the energy gap, the stronger the coupling is. The pure π,π^* state is fluorescent whereas the pure n,π^* state is not and it subtracts molecules from the radiative path through a decay route bypassing the lowest

wherein the coefficients of the linear combination. raised to the 2nd power, i.e. a^2 and b^2 , represent the probability that the molecular system collapses in one of the two states when it is measured. If there exist physical and/or chemical inputs that can alter the values of the coefficients, a and b of equation (2), in continuous manner, an analog phenomenology will appear for any physical quantity dependent on the state represented by ψ_{TOT} . Therefore, a Fuzzy Inference Engine based on the quantum phenomenon ruling the superposition of the two wavefunctions, ψ_1 and ψ_2 , can be built up. ψ_1 esenting the electronic

(3)



excited state (see Fig. 4) [14].

By means of some physical and chemical environmental conditions, such as the temperature (T) and the hydrogen bonding donation (HBD) ability of the solvent, it is possible to influence the extent of interaction between the π,π^* and n,π^* states and hence the values of the coefficients *a* and *b* of the linear combination (equation 3). By measuring the emissive power (Φ_F) of the fluorescent compound, the superposition state Ψ_{TOT} collapses into the $|\Psi_{\pi,\pi^*}\rangle$ state with probability lal² and into $|\Psi_{n,\pi^*}\rangle$ state with probability lbl². The extent of Φ_F is directly proportional to the lal² value. By changing the external inputs, T and HBD of the solvent, in a continuous manner, the *a* and *b* coefficients can assume any real value such that the sum $|a|^2 + |b|^2 = 1$. The final outcome consists in an analog behaviour for Φ_F . This phenomenology, known as Proximity Effect, can be the basis of Fuzzy Logic Systems, wherein the excitation light acts as power supply (see Fig. 5) [15].

Fuzzy Logic Systems can be defined by using the Mamdani's or the Sugeno's methods. Mamdani's method [16] requires that experts perform the fuzzification of the involved input and output variables, by partitioning them in Fuzzy sets, defining the related membership functions (μ) and assigning linguistic variables to each Fuzzy set. IF-THEN statements, inferred by some representative experimental data, are defined as Fuzzy rules. In the case of Proximity Effect, the inputs, T and HBD, and the output, Φ_{F} , are partitioned in a certain number of Fuzzy sets and the multiple antecedents are correlated through the connective AND. Fuzzy rules (R^(j,k)), such as the prototype of equation (4) are formulated:

 $R^{(j,k)}$: IF T is $F^{j}(T)$ AND HBD is $F^{k}(HBD)$, THEN Φ_{F} is $F^{j,k}(\Phi_{F})$ (4)

where F represents a Fuzzy set. Two concrete examples of rules are the following (5) and (6) equations, wherein linguistic variables are associated with each Fuzzy set:

IF HBD is low AND T is warm, THEN Φ_F will be ultralow (5) IF HBD is high AND T is very cold, THEN Φ_F will be veryhigh (6)

As the rules are fixed, the Fuzzy Inference Engine has to be defined and started up. Each Fuzzy rule is interpreted as a Fuzzy implication. Since the antecedent parts of the rules are connected through the AND operator and the cornerstone of scientific modelling, i.e. the cause and effect relation, has to be respected, the membership functions of the rules are defined only by the minimum (equation 7) and the product (equation 8) t-norms:

$\mu_{R^{(j,k)}} = \min[\mu_{F}(T), \mu_{F^{K}}(HBD), \mu_{F^{j,k}}(\Phi_{F})]$	(7)
$\mu_{R^{(j,k)}} = \left[\mu_{F^{j}}(T) \cdot \mu_{F^{k}}(HBD) \cdot \mu_{F^{j,k}}(\Phi_{F}) \right]$	(8)

As the way of determining $\mu_{R(j,k)}$ is fixed, it is necessary to specify how to combine the IF-THEN rules. Generally, they are combined through the t-conorm operator, i.e. the Fuzzy union. The last element of a FLS is the defuzzifier. A criterion for its choice can be based on the attempt of optimising the prediction capabilities of the FLS.

An alternative FLS can be built through the Sugeno's method [17]. It entails rules wherein the output membership functions are either constant or in linear relationship with the inputs (equation 9):

 $R^{(j,k)}$: IF T is F(T) and HBD is $F^{k}(HBD)$, then $\Phi_{F} = p_{(j,k)}F(T) + q_{(j,k)}F^{k}(HBD) + c_{(j,k)}$ (9)

The coefficients $p_{(j,k)}$, $q_{(j,k)}$ and $c_{(j,k)}$ are determined through a Backpropagation-Least-Squares-based Neuro-Fuzzy method and a set of training experimental data. The Sugeno's method guarantees a better description of the Proximity Effect phenomenon.



Other Fuzzy Logic Systems can be implemented by exploiting the behaviour of tryptophan (Trp) upon photo-excitation. The fluorescence quantum yield of Trp and proteins having Trp in their backbone can be modulated through an external physical parameter, such as the temperature and through a chemical compound acting as an effective quencher of Trp's fluorescence, such as flindersine (FL) [18]. An electronically excited Trp can relax through physical paths, such as fluorescence, intersystem crossing, internal conversion and energy transfer to quenching molecules, besides through chemical routes, such as photoionization and intramolecular proton transfer. The last two chemical transformations are thermally activated. Therefore, by increasing the temperature, the energy barrier to non-radiative decay channels can be more easily overcome and the Trp's emissive power weakens. The dependence of Trp fluorescence quantum yield ($\Phi_{\rm E}$) on temperature and molar ratio ($n_{\rm El}/n_{\rm Tro}$) between flindersine (acting as quencher of Trp fluorescence) and Trp itself, is portrayed in Fig. 6.

From the 3D plot it is evident that Φ_F of Trp smoothly changes from high to low values. Φ_F (Trp) is high when both T and n_{FL}/n_{Trp} assume small values. Its dependence on T is stronger at lower temperatures and is attenuated at high values of n_{FL}/n_{Trp} . The gradual lessening of Φ_F (Trp) under the synergetic action of the two inputs, T and n_{FL}/n_{Trp} , is suitable to implement Fuzzy Logic Systems. FLS have been built upon both Mamdani's and Sugeno's method with the multiple antecedents connected through the AND operator (see Fig. 7) [19].

There are also chemical reactions that allow Fuzzy logic to be processed. An example is the biochemical reaction network controlling the glycolysis/gluconeogenesis functions [20]. Here, fructose 6-phosphate (F6P) is interconverted between its two bisphosphate forms by specific kinases and phosphatases. The enzymes in this kinetic mechanism are under the allosteric control of many of the chemical signals of cellular energy status such as cAMP and citrate. The dependence of [F6P] on [cAMP] and [citrate] gives rise to a 3D surface showing a not abrupt transition from low to high values, such as that of Fig. 6. The profile of the 3D surface has a smooth hyperbolic shape and not a steep sigmoidal response: it is apt to process Fuzzy logic.





Fig. 6 - Three-dimensional plot representing the dependence of tryptophan (Trp) fluorescence quantum yield (Φ_F) on temperature (T) and molar ratio of the quencher flindersine (FL) to Trp (nFL/nTrp)

Another example of a Fuzzy chemical reaction is DNA hybridisation wherein two single-stranded DNA molecules (oligonucleotides) bind to form a double stranded DNA duplex. At room temperature, the hybridisation reaction is not a two-state, all or none process, but it is inherently Fuzzy because it is a continuum of outcomes [21]. The pairs of oligonucleotides formed inside a test tube cannot be divided into distinct sets of hybridised and unhybridised species, but each molecule would have a degree of membership in both.

The best implementations of Fuzzy Logic Systems are human senses: sight, hearing, taste, smell and touch are inherently fuzzy. They fuzzify the crisp inputs coming from the outside and send the information to the human brain, that is a Fuzzy inference engine, capable of facing up problems based on subjective or imprecise knowledge. Senses are based on a discrete number of perceiving cells acting as Fuzzy sets [22]. For example, in the case of colour perception, we have three types of cones, whereby we distinguish colours: one cone absorbing mainly the blue portion of the visible spectrum, another absorbing mainly the green and the third principally sensitive to the red. Their absorption spectra in the visible, can be conceived as Fuzzy sets, having Gaussian shape: one centred at 437 nm, the other centred at 533 nm and the third centred at 564 nm. When a radiation, having wavelengths included in the visible, hits the retina of our eyes, it activates the three cones in a specific proportion, i.e. it will have specific values of membership functions in three Fuzzy sets. Each combination for the values of three membership functions will be transduced into the perception of a specific colour inside our brain.

Future perspectives

Molecular computers will probably replace current microelectronic computers. They will be more powerful since information will be

processed at the molecular level and by means of physical and/or chemical inputs and outputs. The hardware of the next molecular computers may be of two types: one that can be defined as "interfacial hardware" and the other that should be dubbed "wetware". In the "interfacial hardware". computations will be carried out by molecules at the interface between a solid and a fluid phase. Molecular logic gates will be anchored to the solid phase and will respond to electrical (for conductive molecules), optical signals or chemical inputs carried by the fluid phase. On the other hand, in the "wetware", soups of suitable chemicals will process information through reaction-diffusion processes mimicking the logic capabilities of cells. Reaction-diffusion computers will have the great advantage of being parallel because myriads of their micro-volumes update their states simultaneously, and molecules diffuse and react in parallel. However, chemical reaction-diffusion computers are supposed to be guite slow if compared with molecular logic gates based upon physical inputs and outputs. Moreover, chemical computers require periodic refuelling and cleaning. This hindrance can be overcome by exploiting non-covalent intermolecular forces as ways of processing information. The resulting supra-molecular species can be disassembled and re-assembled in a reversible way, by modulating the interactions that keep components together.

If the next molecular computers want to fulfil every need of computing, it is compelling to find out how to implement not only hard but also soft computing. In particular, implementation of Fuzzy logic at the molecular level will allow the intelligence quotient of artificial machine to be expanded, up to try to reach the powerful abstraction capability of human reasoning. It will be possible to implement

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Fuzzy Logic Systems through molecules responding to physical inputs in an analog manner, i.e. smoothly, or through chemical inputs if the triggered reactions or supramolecular interactions are a continuum of outcomes. The molecular "interfacial hardware", based on physical and/or chemical inputs and outputs, appears as the sequel of the contemporary computational machineries; the improvements consist mainly in shrinking the logic gates and in the enrichment of signals from merely electrical to electrical/optical and chemical ones. On the other hand the "wetware" strategy appears as a revolutionary computational paradigm, loaded with novelties for artificial intelligence.

Although there is still much work to do before realizing the first binary and Fuzzy molecular and/or chemical computers, the molecular logic devices that are being defined, can find ready application as probes, sensors, and control systems of the microscopic world, useful, also, for healthcare purposes in case they are implanted in living beings.



Pier Luigi Gentili received his PhD in Chemistry from the University of Perugia in 2004. His research activity is focused on the development of Artificial Intelligence by implementing Fuzzy Logic at the molecular level, and Solar Energy's exploitation through hydrogen production by photo-induced water splitting and up conversion of incoherent light.

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Processori molecolari di logica Fuzzy

La tecnologia dell'Informazione è impegnata ad elaborare una nuova generazione di processori: quelli molecolari. Affinché le future macchine artificiali abbiano un elevato quoziente intellettivo, è necessario scoprire com'è possibile elaborare logica Fuzzy su scala molecolare. Infatti la logica Fuzzy consente di trattare le informazioni certe ed incerte, la conoscenza oggettiva e soggettiva: essa permetterà alle macchine di avvicinarsi alla potente capacità di astrazione del ragionamento umano.