

Chemisch kontrollierte Leitfähigkeit: Torsionswinkelabhängigkeit in Biphenyldithiol-Einzelmolekülbruchkontakten**

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Professor Hans Kohn zum 90. Geburtstag gewidmet

Dem Transport von Elektronen durch Moleküle wird heutzutage große Aufmerksamkeit gewidmet, da organische Strukturen als aktive Elemente elektronischer Nanobauteile vorgeschlagen werden.^[1–5] In mehreren experimentellen Anordnungen ist es bereits möglich, einzelne Moleküle in elektronische Schaltkreise zu integrieren und deren Leitfähigkeit zu vermessen. Beispiele sind mechanisch kontrollierte Bruchkontakte (MCBJ)^[6,7] oder molekulare Kontakte auf der Basis von Rastertunnelmikroskopie (STM).^[8–9] die erste grundlegende Untersuchungen der Zusammenhänge zwischen der Struktur und dem Transportverhalten einzelner Moleküle ermöglichten.^[10–14] Es zeigte sich allerdings, dass derartige Experimente von verschiedenen Faktoren abhängig sind. Wichtige solche Faktoren sind 1) die Bildung reproduzierbarer und stabiler Kontakte zwischen beiden Elektroden und dem Molekül, 2) die chemische Struktur und die Konformation des kontaktierten Moleküls und 3) die Algorithmen, die anschließend verwendet werden, um die Messdaten zu verarbeiten. Weil sich mithilfe der chemischen Synthese die Punkte (1) und (2) weitgehend steuern lassen, hat sie eine zentrale Bedeutung erlangt, wenn es gilt, Struktur-Eigenschafts-Beziehungen zu erarbeiten oder maßgeschneiderte Moleküle für elektronische Bauteile zu entwerfen.

Biphenylderivate bestehen aus zwei aromatischen Ringen, die miteinander über eine C-C-Bindung verbunden sind. Sie haben als Modellverbindungen wegen ihrer praktischen Größe und ihrer kommerziellen Verfügbarkeit große

Aufmerksamkeit erlangt.^[15–17] Zusätzlich wurden solche Bausteine als mögliche Schalter diskutiert, weil beide π -Systeme entweder in einer Ebene liegen oder senkrecht aufeinander stehen können („Ein“- bzw. „Aus“-Stellung des molekularen Schalters).^[18–20] Unlängst beschrieben Venkataraman und Mitarbeiter die gegenseitige Abhängigkeit von errechneter Konformation und gemessenem Einzelmolekül-Leitwert in einer Serie unterschiedlich substituierter Biphenyle mit terminalen Aminogruppen.^[21] Trotz mehrerer variierender Faktoren, wie der Elektronendichte in den Phenylringen oder der Größe der Substituenten, wurde ein linearer Zusammenhang zwischen den Leitwerten und dem Quadrat des Cosinus des berechneten Torsionswinkels gefunden. Während solche kurzzeitig immobilisierten Diamine wesentlich zum Verständnis der Struktur-Eigenschafts-Beziehungen beigetragen haben, bilden schwefelfunktionalisierte Strukturen stabilere Elektrode-Molekül-Kontakte.^[22] Auch wurde unlängst vermehrt über Rechnungen an schwefelfunktionalisierten Biphenylen berichtet,^[23–26] und es wurde sogar eine erste Untersuchung an Biphenyldithiol(BPDT)-Kontakten veröffentlicht.^[27] Trotz des großen Interesses an einer Korrelation zwischen Torsionswinkel und Transporteigenschaften konnten bisher keine Verbindungen hergestellt werden, die eine systematische Variation des Torsionswinkels zulassen. Hier stellen wir unseren neuesten Ansatz vor, der die Einstellung des Torsionswinkels in Biphenylsystemen ermöglicht.

Wie in Abbildung 1 dargestellt, sind die 2- und die 2'-Position des Biphenylsystems in den untersuchten Molekülen 1–5 durch Alkylketten unterschiedlicher Länge verbunden, wodurch der Torsionswinkel Φ eingestellt wird. In dieser

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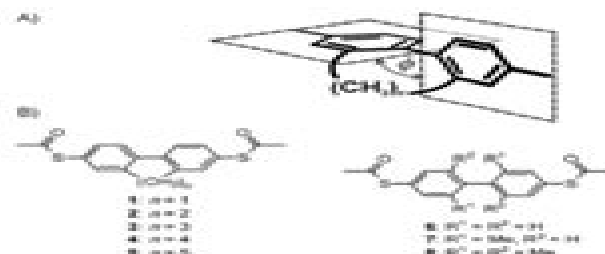


Abbildung 1. A) Der Torsionswinkel Φ zwischen den Ebenen der beiden Phenylringe wird über die Länge der verbindenden Alkylkette eingestellt. B) Die eingesetzten Verbindungen 1–5.

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Volkmar Lankau

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