

$\Delta S < 0$ also means that the latent heat $T_m \Delta S$ is negative, and hence the sample releases heat upon melting on increasing the field, in contrast to the usual melting in which the sample absorbs heat. Note, however, that by crossing the transition lines by increasing the temperature, the latent heat is always positive, as dictated by thermodynamics, and the high-temperature phase always has larger entropy. The observation of a rather constant ΔB in Fig. 4 resolves another previously reported⁴ inconsistency in the behaviour near T_{CP} : at a true critical point ΔB is expected to vanish continuously, whereas experimentally a rather constant ΔB was found to disappear abruptly at T_{CP} . The consistent values of ΔB above and below T_{CP} clearly indicate a smooth continuation of the first-order transition and the absence of a real critical point. However, we emphasize that the vortex system may not be in full thermal equilibrium in the presence of H_{ac} agitation. Therefore the relatively large values of ΔB in Fig. 4, which are found to be sample dependent, should be investigated further.

Finally, an important open question is whether the two disordered states of the vortex matter, liquid and amorphous, represent different thermodynamic phases or just a continuous slowing down of vortex dynamics. Recent numerical calculations led to contradictory conclusions^{25–27,29}. We do not observe any sharp features along the depinning line T_d , which usually extends upwards³⁰ from T_{CP} and is believed to separate the two phases. A strongly first-order T_d transition would require a sharp change in slope of $B_m(T)$ at T_{CP} , which we do not observe. Therefore, the T_d line in BSCCO is unlikely to be a first-order transition; it could be either a continuous transition or a dynamic crossover. Similarly, in $YBa_2Cu_3O_7$ the structure of the phase diagram is controversial, and it is as yet unclear whether the second magnetization peak line merges with the melting line at the critical point¹⁶ or at a lower field. □

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Correspondence and requests for materials should be addressed to N.A. (e-mail: nurit.avraham@weizmann.ac.il).

Coherent transfer of Cooper pairs by a movable grain

L. Y. Gorelik^{*†‡}, A. Isacson^{*†‡}, Y. M. Galperin^{†‡}, R. I. Shekhter^{*†‡} & M. Jonson^{*†‡}

^{*} Department of Applied Physics, Chalmers University of Technology; and Göteborg University, SE-412 96 Göteborg, Sweden
[†] Department of Physics, University of Oslo, PO Box 1048, N-0316 Oslo, Norway; and Division of Solid State Physics, Ioffe Institute of the Russian Academy of Sciences, St Petersburg 194021, Russia
[‡] Centre for Advanced Study, Drammensveien 78, 0271 Oslo, Norway

Superconducting circuits that incorporate Josephson junctions are of considerable experimental and theoretical interest, particularly in the context of quantum computing^{1–5}. A nanometre-sized superconducting grain (commonly referred to as a Cooper-pair box²) connected to a reservoir by a Josephson junction is an important example of such a system. Although the grain contains a large number of electrons, it has been experimentally demonstrated⁶ that its states are given by a superposition of only two charge states (differing by $2e$, where e is the electronic charge). Coupling between charge transfer and mechanical motion in nanometre-sized structures has also received considerable attention^{7–10}. Here we demonstrate theoretically that a movable Cooper-pair box oscillating periodically between two remote superconducting electrodes can serve as a mediator of Josephson coupling, leading to coherent transfer of Cooper pairs between the electrodes. Both the magnitude and the direction of the resulting Josephson current can be controlled by externally applied electrostatic fields.

First let us consider an isolated superconducting grain that is brought into direct contact with a bulk superconductor and then removed. Suppose that initially the grain was in a ground state $|n\rangle$ with a fixed number of Cooper pairs n , counted from the neutral state, while the macroscopic lead, labelled b, was in a ground state

$|\Phi_b\rangle$ with a fixed phase Φ_b of the superconducting condensate. If the switching time δt of the Josephson coupling is long enough to avoid excitations of quasiparticle states ($\delta t \Delta \ll \hbar$, where Δ is the superconducting gap and $\hbar$ is Planck's constant divided by 2π) the interaction between the grain and the lead is reduced to coherent Cooper-pair exchange (Josephson coupling). In this situation the initial state $|n\rangle|\Phi_b\rangle$ evolves into the final state $|\psi_n\rangle|\Phi_b\rangle$ where the state of the grain $|\psi_n\rangle$ can be written (as shown in the Methods):

$$|\psi_n\rangle = \sum_m s_{nm}(\Phi_b)|m\rangle = \sum_m s_{nm}(0)e^{i(n-m)\Phi_b}|m\rangle \quad (1)$$

Below we will refer to such a state as a 'Josephson hybrid'.

An important issue is that a Josephson hybrid can be created in this way only if in the contact position $\mathbf{r} = \mathbf{r}_0$ the differences between the Coulomb energies $E_n = (2en + q(\mathbf{r}))^2/2C(\mathbf{r})$ for a grain with different numbers of Cooper pairs do not exceed the Josephson energy E_J ($\mathbf{r}$ is the grain coordinate, $q(\mathbf{r})$ is the induced charge on the grain and $C(\mathbf{r})$ is the mutual capacitance). Otherwise the Coulomb blockade effect will suppress Cooper pair tunnelling and the grain will remain in the initial state. By applying gate electrodes which control the induced charge on the grain this can be achieved even if E_J tends to zero. If the gate voltage is properly chosen (say, $q(\mathbf{r}) = -e$), the Coulomb energies of the states with two subsequent numbers of Cooper pairs $n = 0, 1$ become equal during the contact time. In other words, the Coulomb blockade of tunnelling is lifted. As a result, the Josephson hybrid consists of only two states, $|0\rangle$ and $|1\rangle$. This is the single-Cooper-pair box that we shall consider. In this case, the final states of the grain $|\psi_{n=0,1}\rangle$ will be given by superpositions $|\psi_0\rangle = s_{00}|0\rangle + s_{10}e^{-i\Phi_b}|1\rangle$ and $|\psi_1\rangle = s_{11}|1\rangle + s_{01}e^{i\Phi_b}|0\rangle$. Because the Cooper-pair exchange between grain and lead decays strongly with distance, we can find the transition amplitudes s_{nm} by considering a situation when the Josephson coupling is instantaneously switched on at time $t = 0$, kept constant during a time interval t_0 (the effective contact time) and then switched off. The state of the grain, initially $|0\rangle$ or $|1\rangle$ at $t < 0$, will then evolve coherently between $|0\rangle$ and $|1\rangle$, giving rise to Rabi oscillations of the two populations with frequency $\Omega_R = \hbar^{-1}E_J$ at $t > 0$. Such oscillations were experimentally observed in ref. 6. Therefore the Josephson-hybrid amplitudes for the isolated grain will depend on the phase of the Rabi oscillations at the end of the contact time interval: $|s_{01}| = |s_{10}| = |\sin\vartheta|$ and $|s_{00}| = |s_{11}| = |\cos\vartheta|$, where $\vartheta = \Omega_R t_0$. Furthermore, a grain initially

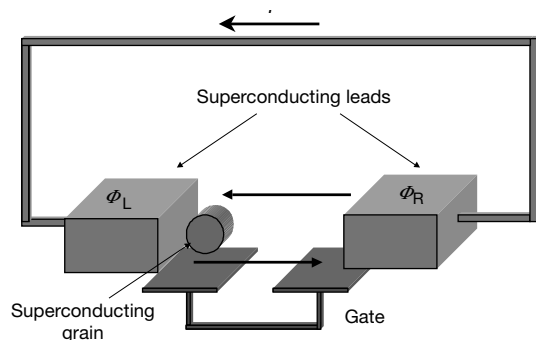


Figure 1 Schematic representation of the nanoelectromechanical system discussed in the text. A superconducting grain executes periodic motion between two superconducting bulk leads connected by a low-inductance superconducting wire. The latter facilitates a fixed phase difference between the leads. The presence of the gate ensures that the Coulomb blockade is lifted during the contacts between the grain and superconductor, which allows the grain to be in a Josephson hybrid state. As the grain moves between the leads Cooper pairs are shuttled between them creating a d.c. current through the structure.

in a hybrid state $|\psi\rangle = \alpha|0\rangle + \beta|1\rangle$ will, according to the superposition principle, evolve into a state $|\psi'\rangle = \alpha|\psi_0\rangle + \beta|\psi_1\rangle$ during such an event.

We now consider a grain that executes periodic motion between two superconducting leads labelled left (L) and right (R), touching them sequentially (Fig. 1). In view of the above discussion it is convenient to describe the state of the grain as $|\psi(t)\rangle = \sum_n c_n(t)e^{i\varphi_n(t)}|n\rangle$. As Josephson tunnelling occurs at each contact the single-Cooper-pair box that is executing periodic motion will mediate an indirect coherent Cooper-pair exchange between the remote leads. The total charge transmitted during a finite interval of time is determined by the whole history of the evolution of the Josephson hybrid and fluctuates strongly in general. Because the Josephson tunnelling is suppressed between the leads, the coefficients c_n remain constant while the phases φ_n evolve according to $\dot{\varphi}_n = \hbar^{-1}E_n$. In general the time evolution of the relative phases $\chi_n = \varphi_{n+1} - \varphi_n$ will depend on n . Leggett and Sols, who first addressed the question of how long the 'memory' of proximity between two remote superconductors can persist, pointed this out as an inevitable intrinsic source of destruction of such a memory¹¹. As only two charge states, characterized by a single relative phase $\chi = \varphi_1 - \varphi_0$, are involved in our Josephson hybrid, there is no room for such dephasing. However, any relaxation that is weak compared to other relevant energy scales (for example, in a real situation relaxation is caused by incoherent charge transfer owing to quasiparticle tunnelling between the grain and the leads) will remove any 'memory' of the initial conditions and the system will evolve into a stationary regime in which the grain state reproduces itself after each period. In this periodic regime we expect directed charge transfer (Josephson current) between the bulk superconductors to appear.

To find such a stationary regime we must consider the evolution of the Josephson hybrid during one period. This evolution can be presented as a sequence of independent events as shown in Fig. 2: (1) Accumulation of relative phase χ while the isolated grain moves between the electrodes. This accumulation is controlled by the electrostatic potential (gate-induced charge $q(\mathbf{r})$) between the electrodes and defined by the equation $\dot{\chi} = \hbar^{-1}(E_1 - E_0)$.

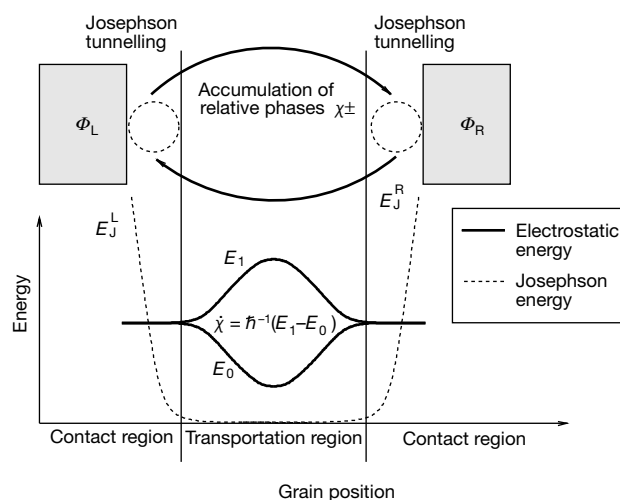


Figure 2 Illustration of the charge transport process. The grain executes periodic motion between the leads. In the contact regions the Josephson couplings $E_J^{L,R}$ are non-zero while the charging energy of the two charge states $|0\rangle$ and $|1\rangle$ are degenerate. In these regions Josephson tunnelling of Cooper pairs take place. As the grain moves into the transportation region the Josephson coupling is suppressed while the degeneracy is lifted. The relative phase χ of the Josephson hybrid now evolves according to $\dot{\chi} = \hbar^{-1}(E_1 - E_0)$. We note that the energy scales of the Josephson energy and the electrostatic energy are not the same.

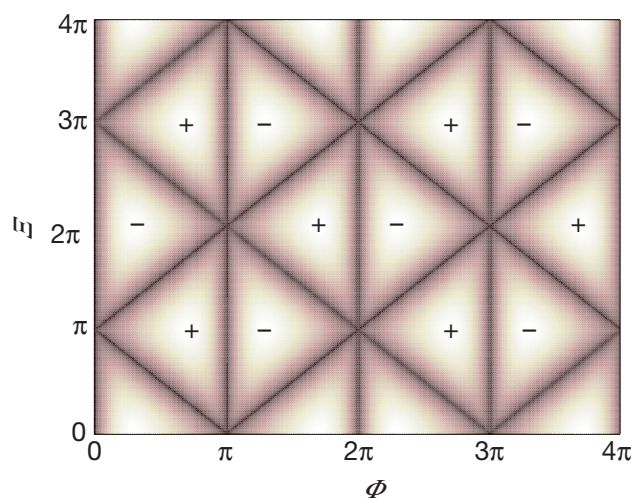


Figure 3 The magnitude of the d.c. current $\bar{I}$ in units of $I_0 = 2ef$ as a function of the phases Φ and Ξ . Regions of black correspond to no current and regions of white to $|\bar{I}/I_0| = 0.5$. The direction of the current is indicated by + or – signs. To see the triangular

structure of the current the contact time has been chosen as $\vartheta = \pi/3$. I_0 contains only the fundamental frequency of the grain's motion and the Cooper-pair charge.

(2) Transitions between states $|0\rangle$ and $|1\rangle$ as a result of Cooper-pair exchange with the bulk superconductors during a finite time interval. The phase of the probability amplitudes for these transitions is controlled by the phases $\Phi_{L,R}$ of the order parameter in the leads, while their moduli s_{nm} are determined by the contact time (see above).

It follows that in a stationary regime the evolution of the Josephson hybrid, and consequently the Josephson current, will be determined by the superconducting phases $\Phi_{L,R}$ as well as by the total relative phases χ_+ (χ_-) accumulated during the motion from the left to the right (right to left) electrode.

To calculate the Josephson current we have found a periodic solution to the quantum Liouville equation for the statistical operator $\hat{\rho}(t)$ of the system. This solution does not depend on initial conditions owing to the presence of weak relaxation caused by incoherent quasiparticle tunnelling. As shown in the Methods, the resulting d.c. current is:

$$\bar{I} = 2ef \frac{\cos\vartheta \sin^3\vartheta \sin\Phi (\cos\Xi + \cos\Phi)}{1 - (\cos^2\vartheta \cos\Xi - \sin^2\vartheta \cos\Phi)^2} \quad (2)$$

This expression contains only the fundamental frequency f of the grain's motion, the Cooper pair charge $2e$, $\Xi \equiv \chi_+ + \chi_-$ and $\Phi \equiv (\Phi_R - \Phi_L) + (\chi_+ - \chi_-)$. How the current depends on the quantities Ξ and Φ is illustrated in Fig. 3.

We observe that the d.c. current is an oscillatory function of the phase difference $\Phi_R - \Phi_L$. This is a direct manifestation of a Josephson coupling between the remote superconductors. We note that the direction of the Josephson current for a given phase difference Φ is controlled by the total relative phase Ξ accumulated during one period, which is determined by the electrostatic gate potential. Furthermore, even when the bulk superconductors have identical phases, $\Phi_R = \Phi_L$, the current does not vanish if the grain's trajectory is asymmetric, $\chi_+ \neq \chi_-$. Also, if the trajectory encloses some magnetic flux created by an external magnetic field with vector potential $\mathbf{A}(\mathbf{r})$, an additional phase $(2\pi/\Phi_0) \oint \mathbf{A}(\mathbf{r}) d\mathbf{r}$ enters the expression for the phase difference Φ , which must be gauge-invariant. Here $\Phi_0 = hc/(2e)$, where c is the speed of light in vacuum, is the magnetic flux quantum.

In the above consideration we assume that the phase difference $\Phi_R - \Phi_L$ as well as the phases $\chi_{\pm}$ remain constant during the observation time. Experimentally, this means that the system should be screened from time-dependent magnetic and electric

fields. The gate electrode, as well as the insulating region between the gate and the grain, should contain as few charged dynamic defects as possible. Time-dependent switching between the states of those defects would produce charge fluctuations coupled to the charge states of the grain. Some residual dephasing should originate from charge fluctuation in the leads during contacts with the grain. The role of low-frequency fluctuations of external fields in inducing slow (compared to the oscillation period) variations of the phases $\Phi_R - \Phi_L$ and $\chi_{\pm}$ can be analysed using equation (2) by assuming the phases to be time-dependent. The current is then obtained by averaging equation (2) over the proper phase distribution. In particular, slow variations of the phase Ξ do not destroy the Cooper-pair pumping, whereas fluctuations of $\Phi_R - \Phi_L$ will suppress the current in the same fashion as in an ordinary Josephson contact.

It seems very difficult to estimate the total dephasing time theoretically. Instead, one can use the experimental results obtained by ref. 6 who observed a coherent response of a superconducting qubit during a time $\tau_{\varphi} = 10^{-9} - 10^{-8}$ s. This is a lower bound for the decoherence time in our case, because the grain is only coupled to the leads during a small part of the period of its motion. Consequently, the contact time is of the order of $\tau_c \approx T/\sqrt{l/d}$ where T is the period of the motion, d is the distance between the leads and l is the characteristic length for Josephson tunnelling, that is, $E_J^{L,R} \approx E_0 \exp(-|\delta\mathbf{r}_{L,R}|/l)$ where $|\delta\mathbf{r}_{L,R}|$ are the distances to the respective leads. It is the quantity τ_c that should be compared to the dephasing time, τ_{φ} , because during the rest of the period the grain is decoupled from the leads. For the upper bound on the dephasing time, $\tau_{\varphi} \approx 10^{-3}$ s was obtained¹² from theoretical estimates for systems fully electrically decoupled from the environment. Although the device should be carefully screened from external fields, having a movable object in the system implies that electrostatic coupling to the gate electrode as well as coupling to external degrees of freedom (phonons, photons) due to irradiation cannot be excluded. If the temperature and the quantity $\hbar\tau_c^{-1}$ are both much smaller than Δ , such coupling does not create quasiparticles and consequently does not contribute to the entropy production in the electronic subsystem. Therefore, decoherence due to particle motion can be taken into account by considering an additional force f affecting the macroscopic motion. This force may produce decoherence if it depends on the charge state $|n\rangle$ of the grain. In this case a relative shift $|\delta x| > l$ in the grain position will cause

discrimination between the different components of the hybrid during contacts with the leads and hence destroy coherence. We have estimated the dephasing time from this mechanism to be of the order of 10^{-8} s. To avoid dephasing from mechanical motion one can imagine another implementation of the Cooper pair shuttling where the alternating couplings between the grain and the leads are controlled by time-dependent potential barriers rather than by mechanical motion. In this case the relevant estimate of τ_φ is obtained from the experiment in ref. 6. $\square$

Methods

Derivation of equation (1)

In the case of Josephson coupling the set of conjugate variables that completely describe the system dynamics³ are $(\hat{\Phi}, \hat{N})$ and $(\hat{\phi}, \hat{n})$. Here $\hat{\Phi}(\hat{\phi})$ and $\hat{N}(\hat{n})$ are the phase and number operators of the lead (grain) condensate. In these variables the interaction between the grain and the lead is $\hat{H}_{\text{int}} = -E_J \cos(\hat{\Phi} - \hat{\phi})$ where E_J is the Josephson energy. As the initial state of the lead $|\Phi_0\rangle$ is an eigenstate of $\hat{H}_{\text{int}}$, we find $|n\rangle|\Phi_0\rangle \rightarrow \sum_m S_{nm}(\Phi_0)|m\rangle|\Phi_0\rangle$. From the commutation relation $[\hat{\Phi}, \hat{N}] = i$ it follows that $e^{-i\Phi_0 \hat{N}}|\Phi = 0\rangle = |\Phi_0\rangle$. Because the interaction conserves the total number of particles, $[\hat{n} + \hat{N}, \hat{H}_{\text{int}}] = 0$, we have $e^{-i\Phi_0 \hat{n}}|n\rangle|\Phi_0\rangle = e^{-i\Phi_0(\hat{n} + \hat{N})}|n\rangle|\Phi = 0\rangle \rightarrow \sum_m S_{nm}(0)e^{-im\Phi_0}|m\rangle|\Phi_0\rangle$.

Derivation of equation (2)

Consider a point $\mathbf{r}_A$ that the grain passes once each cycle, that is, $\mathbf{r}(t_A) = \mathbf{r}(t_A + mT) = \mathbf{r}_A$, $m = 1, 2, 3, \dots$, where T is the period of the motion. The charging term $\hat{H}_C$ and the Josephson term $\hat{H}_J$ in our hamiltonian

$$\hat{H} = \hat{H}_C + \hat{H}_J = (2e\hat{n} + q(\mathbf{r}))^2/2C(\mathbf{r}) - \sum_{s=L,R} E_J^s(\mathbf{r})\cos(\hat{\Phi}_s - \hat{\phi})$$

are never 'active' at the same time (compare Fig. 2); thus the time evolution operator $\hat{U}(t_2; t_1)$ can easily be found along with the states $|\alpha\rangle$ obeying $\hat{U}(t_A + T; t_A)|\alpha\rangle = e^{-i\epsilon_\alpha}|\alpha\rangle$. Owing to dissipation from quasiparticle tunnelling, memory of initial conditions will be lost and the system will settle into a periodic regime that is stable with respect to weak relaxation. In this case $\hat{\rho}(t)$, diagonal in the representation of quasi energy states $\hat{U}(t; t_A)|\alpha\rangle$ and periodic in time with period T , can be found by solving the quantum Liouville equation:

$$d\hat{\rho}/dt = -(i/\hbar)[\hat{H}(t), \hat{\rho}] - \nu(t)[\hat{\rho} - \hat{\rho}_0(t)] \text{ for } \nu(t) = \nu_0 \exp(-|\delta r_{L,R}(t)|/l),$$

where the last term allows for relaxation to the adiabatic equilibrium state $\hat{\rho}_0(t)$. The d.c. current is determined as the average charge delivered to the right lead:

$$\bar{I} = 2e\langle \hat{j} \rangle = \frac{1}{T} \int_T \text{Tr}[\hat{\rho}(t)\hat{O}]dt.$$

The step function $\theta(x(t))$ ensures that only tunnelling to the right lead is taken into account.

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Correspondence and requests should be addressed to A.I. (e-mail: isac@fy.chalmers.se).

The complex nature of superconductivity in MgB_2 as revealed by the reduced total isotope effect

D. G. Hinks*, H. Claus*† & J. D. Jorgensen*

* Materials Science Division, Argonne National Laboratory, Argonne, Illinois 60438, USA

† Department of Physics, University of Illinois at Chicago, Chicago, Illinois 60607, USA

Magnesium diboride, MgB_2 , was recently observed to become superconducting¹ at 39 K, which is the highest known transition temperature for a non-copper-oxide bulk material. Isotope-effect measurements, in which atoms are substituted by isotopes of different mass to systematically change the phonon frequencies, are one of the fundamental tests of the nature of the superconducting mechanism in a material. In a conventional Barden–Cooper–Schrieffer (BCS) superconductor, where the mechanism is mediated by electron–phonon coupling, the total isotope-effect coefficient (in this case, the sum of both the Mg and B coefficients) should be about 0.5. The boron isotope effect was previously shown to be large² and that was sufficient to establish that MgB_2 is a conventional superconductor, but the Mg effect has not hitherto been measured. Here we report the determination of the Mg isotope effect, which is small but measurable. The total reduced isotope-effect coefficient is 0.32, which is much lower than the value expected for a typical BCS superconductor. The low value could be due to complex materials properties, and would seem to require both a large electron–phonon coupling constant and a value of μ^* (the repulsive electron–electron interaction) larger than found for most simple metals.

The search for superconductivity in transition-metal diboride compounds is not new. In 1970, Cooper *et al.*³ were able to obtain transition temperatures (T_c) above 11 K in $\text{Zr}_{0.13}\text{Mo}_{0.87}\text{B}_2$. They also observed superconductivity in NbB_2 (ref. 3). However, as this and other searches⁴ showed, superconductivity in this class of compounds is not common, and finding superconductivity near 39 K in MgB_2 was certainly unexpected.

The presence of an isotope effect is a strong indicator of phonon mediation of the superconducting coupling. The large isotope effect measured by Bud'ko *et al.*² for B ($\alpha_B = 0.26(3)$ where (3) indicates the error in the last decimal place) clearly shows that phonons associated with B vibrations play a significant role in the superconductivity of MgB_2 . The isotope effect coefficient, α , is defined as $\alpha = -\ln T_c / \ln M$ for a one-component system, where M is the atomic mass, and as $\alpha_i = -\ln T_c / \ln M_i$ for each component, i , in a multicomponent system. The MgB_2 system is unique in that both elements have isotopes with isotopic mass differences that could

Table 1 The measured T_c s and calculated α_B s for the isotopic MgB_2 samples

B mass	Mg mass	Onset T_c (K)	10% T_c (K)	α_B onset*	α_B 10%*
10.0051	25.001	40.21(2)	40.16(2)	0.31(1)	0.31(1)
10.9952	25.001	39.06(2)	39.00(2)		
10.0051	24.305	40.23(2)	40.21(2)	0.29(1)	0.30(1)
10.9952	24.305	39.16(2)	39.09(2)		
10.0051	24.001	40.25(2)	40.20(2)	0.30(1)	0.31(1)
10.9952	24.001	39.12(2)	39.06(2)		
Average				0.30(1)	0.31(1)

* α_B is calculated for each of the Mg isotopes for the two T_c criteria.